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An experimental and theoretical investigation of the preferred orientation in deformed and recrystallized hafnium

David S. Gould

AN EXPERIMENTAL AND THEORETICAL INVESTIGATION OF THE PREFERRED ORIENTATION IN DEFORMED AND RECRYSTALLIZED HAFNIUM

An Abstract

of a Dissertation

Presented to

the Faculty of the Graduate School

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In Partial Fulfillment

of the Requirements for the Degree

Doctor of Philosophy

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by

David States Gould

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AN EXPERIMENTAL AND THEORETICAL INVESTIGATION OF THE PREFERRED ORIENTATION IN DEFORMED AND RECRYSTALLIZED HAFNIUM

The deformation and recrystallization characteristics of hafnium containing 3% Zr were studied. The textures of the cold rolled and recrystallized material were determined using an improved Schulz technique which is described. The activation energy for recrystallization was calculated.

The original work employed hafnium crystal bar which was found to be rather brittle. The limit of cold reduction on this material was found to be 25% reduction in thickness. At 500°C., however, the material was found to be more plastic and reductions of approximately 90% in thickness were made. This "warm rolled" sheet material was found to have a (0001) $[10\bar{1}0]$ texture rotated 23° ± 2° in the transverse direction. Compression reductions at 200°C. were made also on samples sectioned from crystal bar. Such samples reduced 90% were found to have an average texture characterized by the basal plane rotated 23° out of the compression plane around a $\langle 10\bar{1}0 \rangle$ rotational axis.

Arc melted hafnium of the same chemical analysis was found to be more ductile. Arc melted samples were cold reduced by rolling 95% without difficulty. Cold rolled strip exhibited a (0001) $[10\bar{1}0]$ texture rotated 25° ± 2° in the transverse direction. The material was then found to have a relatively greater strain hardening tendency than zirconium. The cold rolled texture was rationalized on the basis of the Calnan and Clews method of texture analysis. From a considera-

tion of the deformation mechanisms of titanium and zirconium, which have similar crystal structure and textures, (0001), $(10\bar{1}0)$ and $(10\bar{1}1)$ slip and $\{10\bar{1}2\}$ and $\{11\bar{2}1\}$ type twinning were all assumed to be active. It was shown that these mechanisms could account for the experimentally determined texture. On the basis of crystal structure a comparative analysis of the other hexagonal metals was made and their textures rationalized.

The recrystallized texture of 95% cold reduced 3% Zr arc melted hafnium was found to be approximately (0001) $[11\bar{2}0]$ rotated 5° ± 2° in the transverse direction. Complete recrystallization was found to take place in the above material by annealling one hour at 800°C.

On the basis of change in texture and employing a unique determination technique, the activation energy for recrystallization of 95% cold reduced 3% Zr hafnium was found to be 75. ± 15. K/gm mol.

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ACKNOWLEDGEMENTS

The author would like to thank the metallurgical personnel of the Rolla, Missouri and Albany, Oregon stations of the U. S. Bureau of Mines and the Argonne National Laboratory for making the metallic hafnium used in this investigation available. The author is also grateful to the Los Alamos National Laboratory for making their X-ray diffraction equipment available to him during the summer of 1955.

The author is also indebted to Dr. D. S. Eppelsheimer, Professor of Metallurgical Engineering, for his assistance, support and encouragement during the course of this investigation.

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CHAPTER I

INTRODUCTION

Although not at all a rare element, hafnium has until recent years remained a laboratory curiosity. Hafnium is usually associated with zirconium and hafnium compounds occur naturally with zirconium minerals in appreciable amounts. Hafnium has similar chemical properties and is reduced with metallic zirconium during the production of the later. In that zirconium has not until recently been produced in any significant quantities, the amount of hafnium available from this source has been limited to milligram quantities.

With the emergence of nuclear reactor technology, the practical applications of both zirconium and hafnium have become apparent. Because of the need for hafnium free zirconium, the elements are now being separated on a relatively large scale. As a result appreciable amounts of relatively high grade hafnium have become available for study and limited nuclear application.

For the past several years there has been at the School of Mines and Metallurgy a continuing interest in the deformation mechanisms and resulting textures of the hexagonal metals. It has been felt that if the deformation processes of the hexagonal metals could be understood and the operating mechanisms for a given deformation process predicted, the key to the texture development and deformability of all metals might be developed. This idea of course takes into consideration the fact that the face-centered cubic system is actually a special case of the hexagonal system. Therefore, if an

overall theory could be devised that would work for the above metals, it would encompass all but seven or eight of the engineering metals. This study is only a small part of this rather ambitious project.

For all practical purposes the hexagonal system should be separated into three subsystems; those metals with c/a ratios greater than ideal packing, 1.63, being the first, those with c/a at or very near 1.63 being the second and those with c/a ratios less than 1.63 being the third. Metals such as zinc and cadmium would exemplify the first group, magnesium the second and titanium, zirconium and hafnium the third. The textures of magnesium and zinc have been rather intensely covered, but prior to 1951 there was little or no reliable reported literature on deformation studies of the metals with c/a ratios of less than 1.63, the third group above. This group, therefore, became the primary field of study for those interested in completing the overall study of the hexagonal metals. Titanium was investigated at the School of Mines and Metallurgy; zirconium and beryllium elsewhere. In 1954 a small amount of hafnium crystal bar became available and this predicated the investigation of hafnium.

Through the years most hexagonal metals, notably magnesium and zinc, have been considered as being very difficult to cold work. This concept has spread to the point that all hexagonal metals are more or less considered to be unsuitable for cold working. It has been shown that titanium and zirconium will undergo plastic deformation without any insurmountable trouble and are capable of being cold worked to an extent beyond most other hexagonal metals. It was therefore of

interest to see if this relatively recent discovery could be extended to hafnium as well. If hafnium could be cold worked, its application in the nuclear energy field would be greatly simplified as a result of easier fabrication techniques.

The research problem, as presented, therefore became twofold; first to investigate and determine the feasibility of fabrication of hafnium and secondly to study the deformation characteristics of the material in light of the overall study of hexagonal metals aimed at the development of an overall general theory of plastic deformation. This appeared to envolve a study of the operating deformation systems in hafnium and the theoretical and experimental study of the textures resulting from plastic deformation. These were to be analyzed in comparison with the other hexagonal metals.

It became evident immediately that the research techniques which must be evolved for the study of the textures of hafnium might have considerable application in subsequent work. Therefore the development of texture determination procedures using relatively refined equipment was considered part of the research problem.

CHAPTER II

REVIEW OF THE LITERATURE

In that the present work is part of a study of the deformation characteristics of the whole hexagonal system, the reader is referred to the previous published work of Williams and Eppelsheimer in this general field [1, 2, 3, 4]. In this series of articles the early work on texture determination and the theories of the "German School" first trying to explain plasticity of metals are thoroughly presented. The following survey will be primarily limited to presenting the limited literature relative to the physical properties of hafnium and to a discussion of the newer techniques of texture determination.

Literature about the physical properties of hafnium. At the time the present study was initiated in 1954 the most inclusive article on the physical properties of hafnium was by F. B. Litton [5]. This study investigated the malleability of hafnium, using crystal

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(1) D. N. Williams and D. S. Eppelsheimer, "The cold rolled texture of titanium," Trans., AIME, Vol. 203, p. 1378, 1953.

(2) D. N. Williams and D. S. Eppelsheimer, "Compression texture of iodide titanium," J. of Metals, AIME, Vol. 4, p. 615, 1952.

(3) D. N. Williams and D. S. Eppelsheimer, "A theoretical investigation of the deformation textures of titanium," J. of Inst. of Metals, Vol. 81, p. 553, 1952-53.

(4) D. N. Williams, "Deformation textures of titanium," U. of Mo., Ph.D. Dissertation, (M.S.M.-T1036), 1952.

(5) F. B. Litton, "Preparation and some properties of hafnium metal," J. of Electrochem. Soc., Vol. 98, p. 488 (Dec.), 1951.

bar and arc melted samples. This work indicated that hafnium was harder and more susceptible to strain hardening than zirconium. The limit of plastic deformation by cold rolling was reported as being 65%. No texture studies were included although the lattice parameters reported in this work were subsequently found to be the most applicable in the present study. The eighteen references cited by Litton constitute a good survey of the background of the production of hafnium.

During 1951 the Albany, Oregon Station of the U. S. Bureau of Mines started pilot production and fabrication of hafnium. Although their report of 1955 (6) provides no data about the physical properties of the material produced, the bibliography does provide an outline of the extraction and zirconium separation processes in use to that date.

A microscopic study of the cold worked and annealed hafnium was reported by H. P. Roth of M.I.T. (7). This work noted the similarity of its metallography to that of zirconium. This report also indicated that the recrystallization temperature may be higher than the 700° to 800°C reported by Litton.

The Westinghouse Electric Company was also apparently working with hafnium. One article described a hot swagging process for fabri-

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(6) H. P. Holmes, M. M. Barr and H. L. Gilbert, "Production of hafnium," U. S. Bureau of Mines, Report of Investigations 5169, November, 1955.

(7) H. P. Roth, "Metallography of Hafnium," Metals Progress, ASM, Vol. 63, (June), 1953.

cation of crystal bar[8]. None of their other work is reported in the literature. Private inquiry indicated that this company is doing no fundamental texture work.

The general physical properties of hafnium at different hot and cold work levels was reported by Adenstedt[9]. This was of no more than general interest as far as the present work is concerned.

Generally speaking the reported work on the physical properties of hafnium is outstandingly incomplete. This is undoubtedly due to the fact that until very recently only very small amounts of the material have been available for study. All the reported data, including this study, have been based on something less than ten pounds of metallic hafnium!

Several recent articles explain more fully the newer processes for the separation and reduction of hafnium. In an Atomic Energy Commission report, J. M. Googin describes the two liquid-liquid extraction processes used for the separation of hafnium from zirconium[10]. In several recent articles the reduction and purification of metallic hafnium is discussed[11, 12].

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(8) R. B. Stermon, "Evaluation of hafnium crystal bar," Atomic Energy Commission Report: WAPD-TD-521, August, 1955.

(9) H. K. Adenstedt, "Physical, thermal and electrical properties of hafnium and high purity zirconium," Trans., ASM, Vol. 44, p. 949, 1952.

(10) J. M. Googin, "The large scale separation of zirconium and hafnium," U. S. Atomic Energy Comm. Report Y-B65-103, Sept. 1956.

(11) S. M. Shelton, E. D. Dilling and J. H. McClain, "The production of zirconium and hafnium," Progress in Nuclear Engineering, Series V, Metallurgy and Fuels, Vol. 1, pp. 305-351, 1956.

(12) O. N. Carlson, F. A. Schmidt and H. A. Wilhelm, "Preparation of zirconium and hafnium by bomb reduction of their fluorides," J. of Electrochem. Soc., Vol. 104, p. 51 (Jan.) 1957.

Since titanium, zirconium and hafnium are so similar in properties and behavior; it was advisable to review the literature since 1952 relative to the deformation characteristics of titanium and zirconium.

A recent Russian work by Sokolov, Zaleskiy and Yelyutin notes the resistance to deformation of titanium at various temperatures. Using constant rates of deformation, the resistance to deformation was found to fall off rapidly at temperatures above 700°C [13].

Three articles noted and discussed in Chapter V [14, 15, 16] appear to establish the slip and twinning systems which constitute to deformation mechanisms for titanium.

Three other articles published since Williams and Eppelsheimer's work in 1952-53 definitely establish the cold rolled textures of

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(13) L. N. Sokolov, V. I. Zaleskiy and V. P. Yelyutin, "Resistance to deformation of commercial titanium," Sb. Mosk, in-ta stali, No. 33, p. 143, 1955.

(14) F. D. Rosi, C. A. Dube and B. H. Alexander, "Mechanism of Plastic Flow in Titanium - Determination of Slip and Twinning Elements," J. of Metals, AIME, Vol. 5, p. 257, (Feb.), 1953.

(15) T. S. Liu and M. A. Steinberg, "Twinning in Single Crystals of Titanium," J. of Metals, AIME, Vol. 4, p. 1043, (Oct.), 1952.

(16) E. A. Anderson, D. C. Jillson, and S. R. Dunbar, "Deformation Mechanisms in Alpha Titanium," J. of Metals, AIME, Vol. 5, p. 1191, (Sept.), 1953.

titanium and zirconium [17, 18, 19]. It appears that metals in this group all have cold rolled textures which can be described as (0001) $[10\bar{1}0]$ textures rotated at various angles in the transverse direction.

It is also interesting to note that the duplex structure observed by Williams and Eppelsheimer [20] and attributed to the effect of impurities has also been observed in Ti-Al alloys [21].

X-Ray technique for determination of textures. The basic Schulz-Decker described by Williams [22] in Appendix I of his thesis is still basis of the newer preferred orientation procedures. The recent changes are in the form of refinements with no basic alteration of principles involved.

The basic preferred orientation fixture, which in most cases

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(17) J. H. Keeler and A. G. Beisler, "Preferred orientations in rolled and annealed titanium," J. of Metals, AIME, Vol. 7, p. 80, (Feb.), 1956.

(18) J. H. Keeler, W. R. Hibbard, Jr., and B. F. Decker, "Textures of rolled and annealed zirconium," Trans. AIME, Vol. 197, p. 932, 1953.

(19) R. K. McGeary and B. Lustman, "Kinetics of thermal reorientations in cold rolled zirconium," J. of Metals, AIME, Vol. 5, p. 284, (Feb.), 1953.

(20) Williams and Eppelsheimer, Loc. cit., (Ref. 1).

(21) C. J. Sparks, Jr., C. J. McHargue and J. P. Hammond, "Effects of aluminum on the cold rolled textures of titanium," J. of Metals, AIME, Vol. 9, p. 49, (Jan.), 1957.

(22) Williams, Loc. cit., (Ref. 4).

was similar to the one devised by Williams and Eppelsheimer[23], has evolved into more refined apparatus in several laboratories working more or less independently. Three commercially produced models have been recently marketed; one by General Electric Company[24], one by North American Philips Company[25], and one by Siemens[26] in Germany. The GE fixture uses a transmission technique, while the Norelco and Siemens fixtures use reflection. All are motorized so that the movement of the sample is continuous, providing continuous readings at different positions on the pole figure. The concept of spiral scanning has been introduced which evolves two simultaneous planes of movement and allows the whole pole figure to be covered without need for interruption or adjustment[27]. The General Electric fixture has

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(23) D. N. Williams and D. S. Eppelsheimer, Universal specimen mount for pole figure determination using Schulz-Decker technique," Rev. Sci. Instruments, Vol. 23, pp. 229-231, 1952.

(24) A. H. Geisler, "Crystal orientation and pole-figure determination," GE Research Laboratory Report No. RL-790, Jan., 1953, General Electric Co., Schenectady, N. Y.

(25) W. P. Chernock and D. Wahl, "A preferred orientation specimen holder for use with the Norelco diffractometer," Norelco Reporter, Vol. II, p. 71 (Sep.-Oct.) 1955, North American Philips Company, Mount Vernon, N. Y.

(26) Hans Neff, "A new countergoniometer for the determination of textures in sheets," Z. fur Metallkunde, Vol. 47, p. 646, 1956.

(27) A. N. Holder, "A spiral scanning X-ray reflection goniometer for the rapid determination of preferred orientations," Rev. Sci. Inst., Vol. 24, p. 10, 1953.

even been fitted with an automatic recorder which plots the pole figure as the specimen moves[28]. All of these fixtures have provisions so that the sample moves continuously in its own plane thus providing diffracted intensities which are representative of a chosen area.

Several techniques have been devised to overcome the limited area of the pole figure which can be covered using reflection or transmission using one sample based on one crystallographic face. In the conventional technique only the outer portion of the pole figure can be covered by transmission and only the inner area by reflection. These can be combined to provide complete coverage but sometimes this is impractical or impossible. Mueller and Knott have proposed a technique for sheet material where the inner portion is first covered; the sheet is then sliced into strips which are turned 90° about an axis in the rolling direction and transverse direction, bolted back together and rerun for the outer areas of the pole figure[29]. Another method with the same objective using one unsectioned sample, employs low order diffraction planes in the outer region of the pole figure and adjusts the intensities of the peaks obtained to another

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(28) A. H. Geisler, "Automatic pole-figure recorder," GE Research Laboratory Report No. RL-963, October, 1953, General Electric Company, Schenectady, N. Y.

(29) M. H. Mueller and H. W. Knott, "Quantitative Pole Figures for sheet materials by the reflection technique," Rev. Sci. Inst., Vol. 25, p. 1115, 1954.

set at intensity values obtained from higher order planes for the center of the pole figure[30].

There have also been a number of articles relative to the elimination of experimental error. Chernock and Beck have pointed out the necessity of making tilting angle corrections for the outer area of the reflection pole figure[31]. This procedure is treated in detail in the appendix. Geisler has discussed the problem of spurious areas occurring in the pole figures[32]. This is a problem in titanium and hafnium, in that the (0002) and (10$\bar{1}$1) planes are only about one and one-half degrees apart in 2θ angle. As a result of poor resolution (10$\bar{1}$1) peaks sometimes tend to produce false peaks in (0002) pole figures. Geisler indicates these can be lessened by means of tighter slit systems. Such spurious peaks can be identified by checking filtered photographic preferred orientation determinations where the true nature of the peaks will be obvious and by geometric analysis of all the pole figures obtained for a given sample. In such an analysis the peaks which are crystallographically associated with every other peak in each pole figure can be identified. Spurious

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(30) M. Schwartz, "Method for obtaining complete quantitative pole-figures for flat sheets using one sample and one sample holder," J. Appl. Phys., Vol. 26, p. 1507, 1955.

(31) W. P. Chernock and P. A. Beck, "Analysis of certain errors in the X-ray reflection method for the quantitative determinations of preferred orientations," J. of Appl. Phys., Vol. 23, p. 344, 1952.

(32) A. H. Geisler, "Spurious Areas in Pole figures," GE Research Laboratory Report No. RL-971, November 1953, General Electric Company, Schenectady, N. Y.

peaks in the (0002) pole figure, for instance, will have no associated peaks in the other pole figures. Another important article by Braybrook and Calnan examined the effect of sample surface condition on the resulting diffracted intensity values[33]. They indicated that the ultimate texture determined appeared to be dependent on the amount of surface material removed and the method or methods of removal. They deduced that the most satisfactory procedure for reflection determinations was to remove a minimum of 0.001" of material from the surface by mechanical polishing followed by chemical etching. This happily coincided with the procedure already used by the author.

Origin of textures. The most practical approach for rationalizing the textures developed in cold rolled hexagonal metals, proposed to date, is according to a method explained by Calnan and Clews in a series of three articles[34, 35, 36]. The basic assumption of the

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(33) R. F. Braybrook and E. A. Calnan, "Some observations on the development of face-centered cubic rolling textures," J. Inst. of Metals, Vol. 85, p. 11, (Sept.), 1956.

(34) E. A. Calnan and C. J. B. Clews, "The development of textures in metals - Face-centered cubic system," Phil. Mag., Vol. 41, p. 1085, 1950.

(35) E. A. Calnan and C. J. B. Clews, "The development of textures in metals - Body-centered cubic system," Phil. Mag., Vol. 42, p. 616, 1951.

(36) E. A. Calnan and C. J. B. Clews, "The development of textures in metals - Hexagonal C.P. System," Phil. Mag., Vol. 43, p. 93, 1952.

Calnan and Clews analysis, different from the basis of most others[37], is that in order to maintain grain boundary cohesion, slip need not necessarily occur immediately upon the applied resolved shear stress attaining the critical value for the most favorably oriented slip system. It is assumed that this is due to neighboring grains causing constraints which cause the effective stress to be moved to a slightly different value such that the critical value is not reached. The effective stress is therefore thought to move to lesser resolved shear stress values by a direct route bringing about the possibility of duplex and multiple slip. By analysis of the associated rotations the texture can be deduced. The theory is discussed further in chapter V.

The Calnan and Clews system has been applied to hexagonal metals with notable success.

The only other theoretical proposal published since 1952 relative to the origin of texture in cold-worked metals was a paper presented at British Institute of Physics Conference by T. L. Richards and D. E. Yeomans[38]. This paper extended the theory of the "Unextended Cone" to cubic metals. It assumes a three dimensional "garden trellis" with slip along the [100] direction. This theory predicts a (110) [1$\bar{1}$2] texture for cubic metals. The authors claim a very simple

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(37) G. I. Taylor, "Mechanism of plastic deformation of crystals," Proc. Royal Soc., (London), Vol. 145, pp. 362-404, 1934.

(38) "Summarized proceedings of a conference on the structures and textures of metals - London, October, 1954, "British J. of Applied Physics, Vol. 6, p. 377, (Nov.), 1955.

method of analysis but unfortunately it does not seem to apply to hexagonal metals and there is substantial evidence that even in cubic metals the mechanism is much more complicated. It appears that the true texture for cubic metals may have a true texture closer to a (123) $[\bar{1}\,\bar{2}\,1]$ preferred orientation.

At this same conference Calnan explained and defended his method which, although it has some faults, appears to be the only present method of analysis which can reationalize the texture of cold worked hexagonal metals.

CHAPTER III

DEFORMATION STUDIES OF HAFNIUM CRYSTAL BAR

Several pieces of hafnium crystal bar were supplied to be used for the basis of the deformation study; one from the Argonne National Laboratory and one from the U. S. Bureau of Mines, Albany, Oregon Station. Since previous workers (39) studying titanium had been able to satisfactorily cold work iodide crystal bar, it was assumed that hafnium crystal bar could also be satisfactorily cold worked in this investigation.

A. PROPERTIES OF ORIGINAL CRYSTAL BAR MATERIAL

The physical appearance of one piece of the hafnium crystal bar is shown in Figure 1. It was shiny, dense and appeared to be perfectly formed material with no voids or imperfections.

The etched cross-section of the bar is shown in Figure 2. The hafnium wire on which the iodide material was deposited can be seen in the center surrounded by a layer of very fine grain material, subsequently surrounded by a thicker layer of very coarse grained material.

The two pieces obtained had essentially the same appearance. The chemistry data supplied is shown in Table No. 1. Both samples were Kroll process sponge refined by the iodide process. A sample

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(39) Williams and Eppelsheimer, Loc. cit., (Ref. 2).

FIG. NO. 1

HAFNIUM CRYSTAL BAR

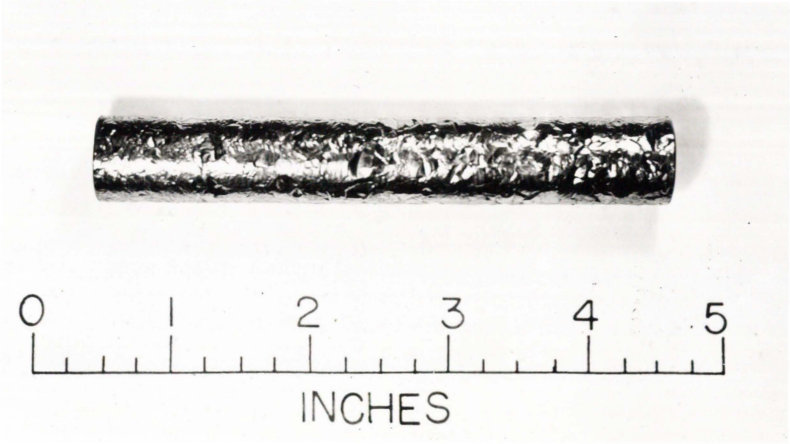

FIG. NO. 2

ETCHED CROSS SECTION OF HAFNIUM CRYSTAL BAR

MAGNIFICATION: APPROXIMATELY 7X

ETCHING SOLUTION: 10% HCL, 10% HNO_3, 1% HF, 79% H_2O

TABLE I

CHEMISTRY OF SAMPLE SUPPLIED BY ARGONNE NATIONAL LABORATORY

Ag - L1	Cr - 2	Ni - 6
Al - 20	Cu - 3	P - L50
As - L10	Fe - 100	Pb - 1
B - 0.6	K - L20	Sb - L1
Ba - L50	Li - L1	Si - L50
Be - 10.2	Mg - 1	Sn - L5
Bi - L1	Mn - 1	Sr - L100
Ca - L100	Mo - L20	Ti - L50
Co - L5	Na - L10	V - L20
Results are expressed in PPM		Zn - L50
L = Less than		Zr - 2-4%

CHEMISTRY OF SAMPLE SUPPLIED BY BUREAU OF MINES

Al - 20	Mg - 40	Ti - 100
Cr - L30	Mo - 60	V - 40
Cu - 40	Ni - 40	Zr - 3%
Fe - 800	Si - 150	O - 120
Results are expressed in PPM		
L = Less than		

of very low zirconium content crystal bar hafnium was also obtained but was not used in the investigation reported herein.

Before sectioning the bar and starting attempts at deforming any sample from it, it was felt necessary to check the orientation of the grains of the bar in the "as produced" condition. A random original orientation is normally assumed in making texture studies. It has been reported that evaporated and deposited films of hexagonal metals have a $[\bar{0}001]$ orientation[40]. It seemed possible therefore that such an orientation might also exist normal to the axis of the crystal bar.

A series of Laue back reflection determinations of individual grains was made. The X-ray beam was adjusted so that it was normal to the axis of the crystal and focused on an individual grain. Two representative pictures are shown in the following figures. The first shows a grain with an orientation very near to $[\bar{0}001]$, the second is tilted of at an angle of approximately 40°. Only 10% of the grains examined showed the predicted $[\bar{0}001]$ direction. On the basis of the back reflection data it would appear that individual crystals do not always assume the $[\bar{0}001]$ preferential growth direction.

The lack of orientation can possibly be explained by the fact that the crystals bars were formed at a temperature in the range of the transformation temperature for hafnium from HCC to FCC[41]. This

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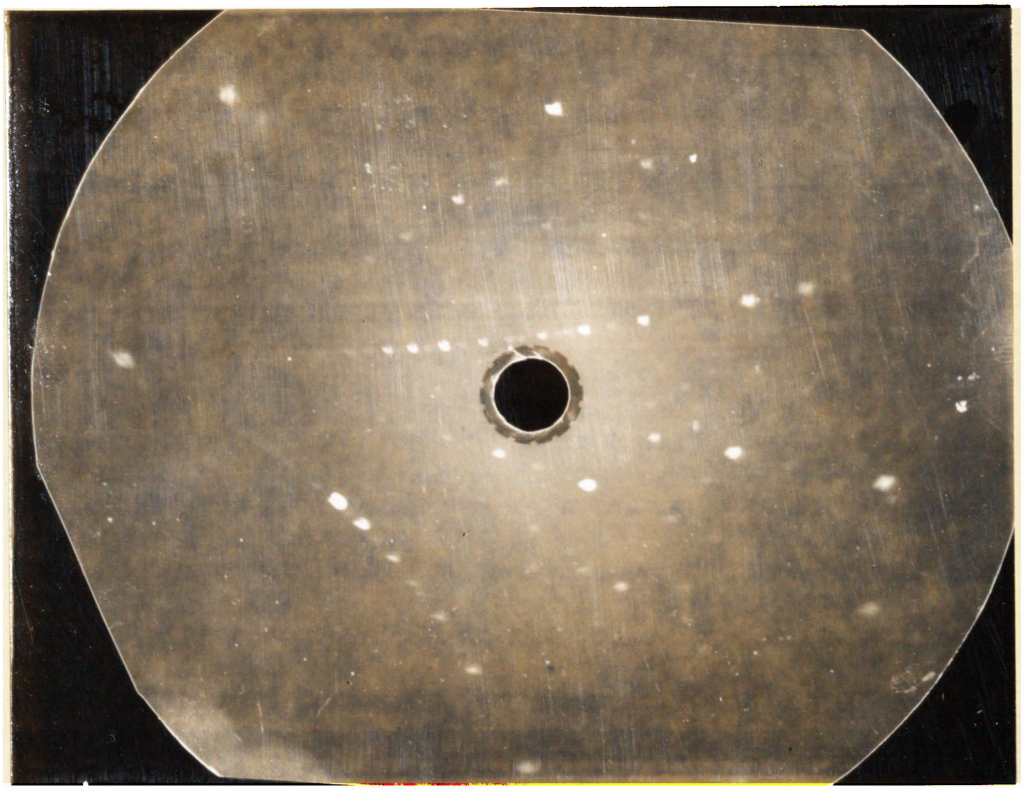
(40) C. S. Barrett, Structure of Metals, (New York, McGraw-Hill Book Company, 1952), p. 519.

(41) Litton, Loc. cit.

FIG. NO. 4

LAUE BACK REFLECTION X RAY PHOTO OF HAFNIUM SINGLE GRAIN

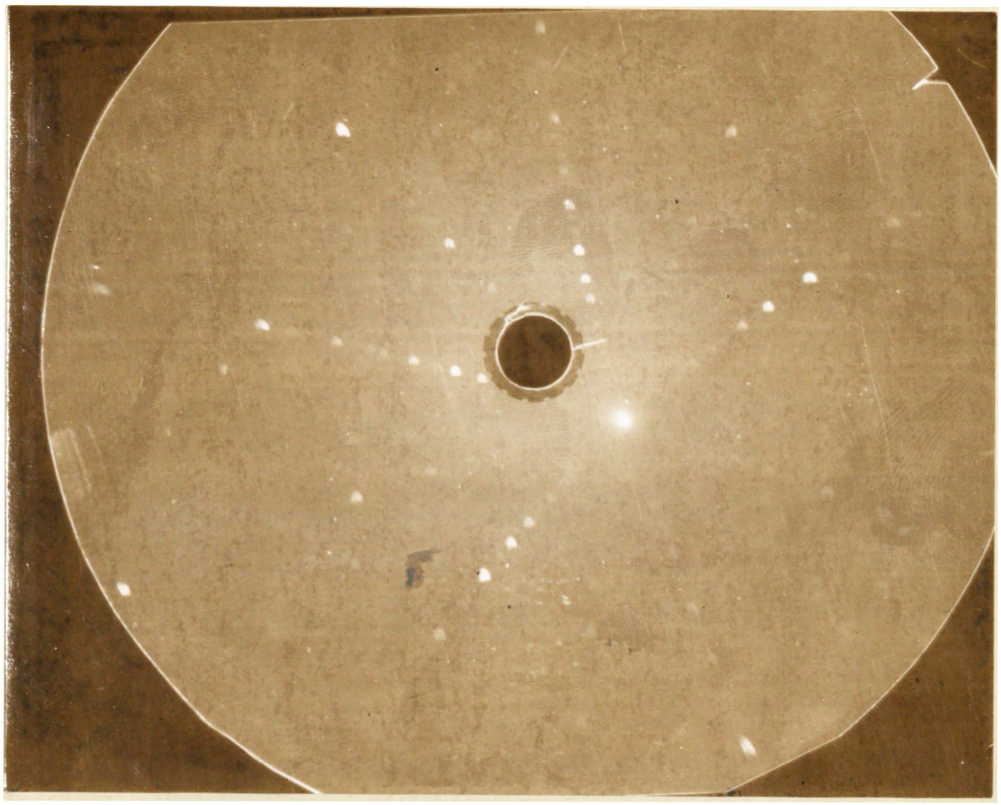


X RAY BEAM NORMAL TO PLANE OF PAPER

X RAY BEAM NORMAL TO AXIS OF CRYSTAL BAR

FIG. NO. 3

LAUE BACK REFLECTION X RAY PHOTO
OF HAFNIUM SINGLE GRAIN



X RAY BEAM NORMAL TO PLANE OF PAPER

X RAY BEAM NORMAL TO AXIS OF CRYSTAL BAR

would indicate that the original crystals might have been FCC originally and transformed to HCP subsequent to their deposition. This idea is further substantiated by the fact that the individual grains appear, upon visual examination, to be pseudomorphs. They appear to be tetragonal or cubic but not hexagonal.

B. COLD DEFORMATION OF CRYSTAL BAR SAMPLES

Transverse sections were cut from the original crystal bar. These cuts were made normal to the longitudinal axis of the bar. When these samples were cold worked, even small amounts, they tended to crack around the original wire core of the bar. It appeared that the bond between the deposited bar material and the wire core was not strong enough to support any plastic deformation.

A second attempt was made to section the bar, slicing the bar down the middle along the longitudinal axis in such a manner that the core material was completely removed. These samples were a little more plastic but cracking was found to originate along the grain boundaries of the very fine inner grains of the original crystal bar after only ten to fifteen percent reduction in thickness. These cracks proceeded through the coarse grained material and caused it to fail at the above relatively low reductions. Subsequently all the fine grain material was milled off similar samples and they were used for deformation studies. These samples were then essentially "Slabs" off the periphery of the crystal bars.

It was later found that even these samples, which consisted of a relatively few very large grains, could only be cold reduced approxi-

mately twenty-five percent before failure occurred at grain boundaries.

In the hope that annealing might improve the cold workability of the samples several samples were heated in vacuum for one hour at 800°C before deformation. Several samples that had been cold worked a very small amount, less than 10% reduction with no cracking, were also annealed with the above. The vacuum of the annealing retort furnace was of the order of 20-30 microns. In both cases there was no evidence that the annealing treatment had any significant effect on the subsequent cold workability of the sample as they cracked at about the same level of cold reduction as the unannealed samples. This seemed to indicate a relatively low strength between grains, which may have stemmed from the original deposition mechanism.

It was decided to determine if the materials could be deformed at temperatures slightly above room temperature. It is generally conceded that metals are more plastic with increasing temperature. Such a process is called "Warm Rolling", to distinguish it from hot rolling which is deformation above the recrystallization temperature. The recrystallization temperature of hafnium was reported to lie between 700° to 800°C[42]. Several deformation runs were attempted at temperatures ranging from 200° to 500°C. At 500°C it was found that the "Slab" samples could be rolled to any practical reduction and that at only 200°C it was possible to achieve compressive reductions of better than ninety percent.

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(42) Litton, Loc. cit.

C. COMPRESSION TEXTURE

Several cylindrically shaped samples were sectioned from the outer portion of the crystal bar material. These samples were approximately 1/4" in diameter and 1/4" high. They were shaped so that the compression axis was the cylinder axis of the sample. The samples contained none of the very fine grain inner material.

The samples were compressed in a 1" diameter metallurgical mounting die using a 60,000 pound capacity testing machine. The die and sample were heated to 300°C before pressure was applied. The temperature was measured by a thermocouple in the mounting die. The compression surfaces of the samples were lubricated by a heavy silicone lubricant. When the testing temperature was reached the testing machine was loaded to full capacity and held for several seconds. The sample was then removed and usually found to be of the order of 1/2" in diameter after compression. This would indicate that the deformation had proceeded at compressive stresses of 300,000 psi or less. The samples were positioned so they were in contact with the compressive surfaces only.

After the first run the periphery of the compressed sample was ground off, reducing the sample again to a 1/4" diameter cylinder. The preceding process was repeated several times. After the sample reached a thickness of approximately 0.040", it was found the subsequent runs provided little or no additional reduction in thickness. The total reduction possible by this process was, therefore, limited by the thickness of the initial samples.

A number of samples were prepared, some with the cylinder axis parallel to the longitudinal axis of the crystal bar and some with the cylinder axis normal to it.

The final samples were disks approximately 0.040" thick and 1/2" in diameter. These samples were ideal for reflection preferred orientation techniques, being a size ideally suited for the stage of the pole figure determination device.

Approximately 0.002" was etched from the surface of each sample and the (0002) and ($10\bar{1}1$) pole figures were run for each sample according to the procedure indicated in the appendix.

The pole figures for three samples which were run according to the conventional technique are shown on the following six pages.

It can be noted that the basal planes are rotated out of the compression plane at various angles from 15° to 30°. In comparing the (0002) and ($10\bar{1}1$) pole figures a very definite preference for a $[\bar{1}010]$ rotational axis can be noted. This generally is in agreement with the findings of Williams and Eppelsheimer for titanium[43], in that they noted a similar rotation angle with a tendency toward a $[\bar{1}010]$ rotational axis.

Following these determinations an integrated determination was made for each sample. This was obtained by rotating the sample rapidly in its own plane, α rotation, while the sample is moved slowly through the ϕ angle. This, in effect, combines the spots shown in the previous pole figures into circular orbits. In this determination the

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(43) Williams and Eppelsheimer, Loc. cit., (Ref. 2).

FIG. NO. 5

INNER 60° PORTION OF

(0002) POLE FIGURE

HAFNIUM CRYSTAL BAR COMPRESSION SAMPLE

STRESS AXIS PARALLEL TO BAR AXIS

76.5 % REDUCTION IN THICKNESS AT 300° C.

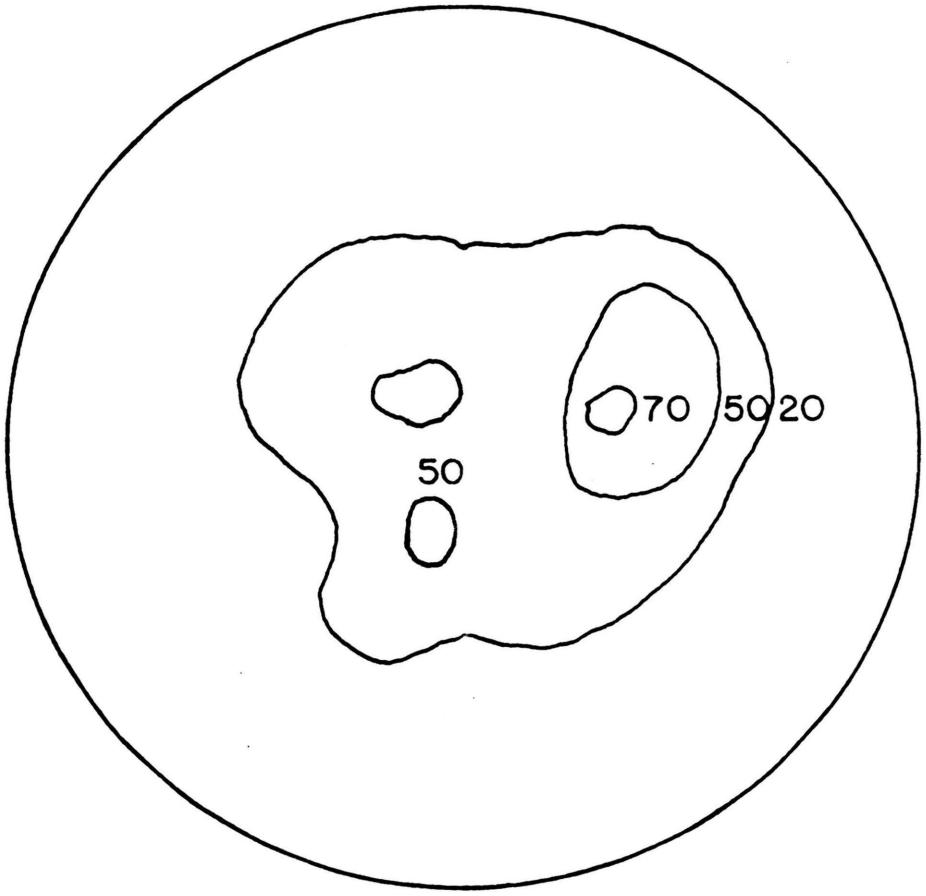


FIG. NO. 6

INNER 60° PORTION OF

(10$\bar{1}$1) POLE FIGURE

HAFNIUM CRYSTAL BAR COMPRESSION SAMPLE

STRESS AXIS PARALLEL TO BAR AXIS

76.5% REDUCTION IN THICKNESS AT 300° C.

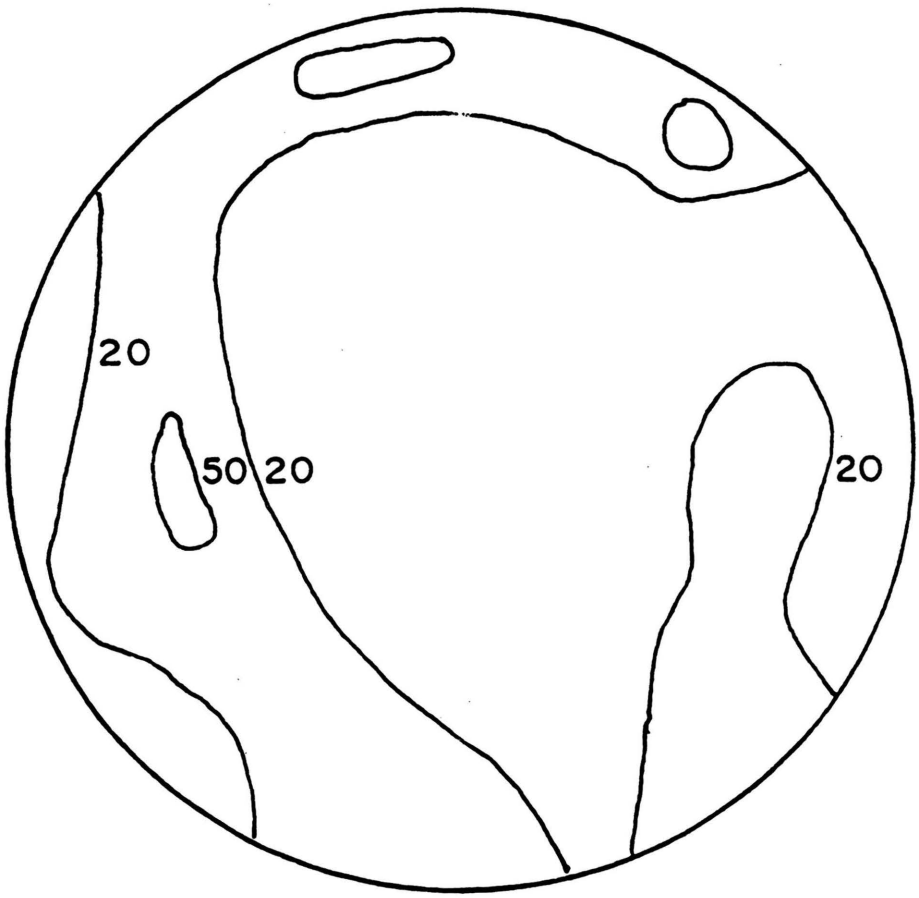


FIG. NO. 7

INNER 60° PORTION OF

(0002) POLE FIGURE

HAFNIUM CRYSTAL BAR COMPRESSION SAMPLE

STRESS AXIS NORMAL TO BAR AXIS

78.1 % REDUCTION IN THICKNESS AT 300° C.

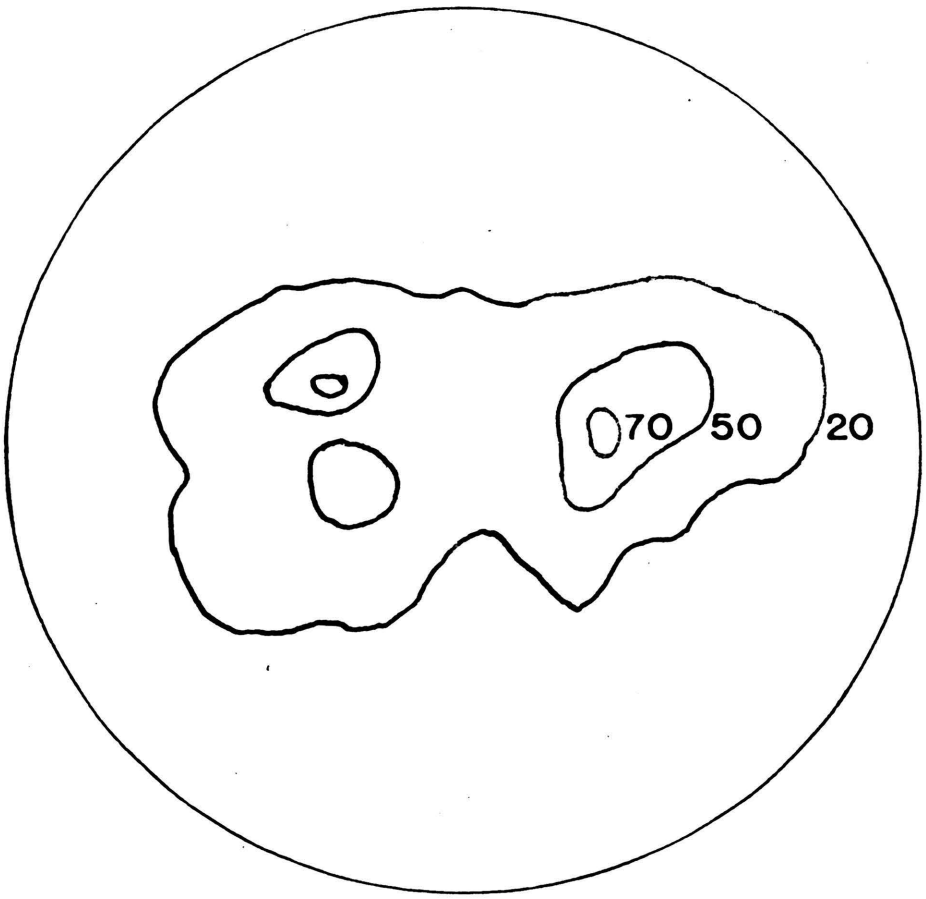


FIG. NO. 8

INNER 60° PORTION OF

(10$\bar{1}$1) POLE FIGURE

HAFNIUM CRYSTAL BAR COMPRESSION SAMPLE

STRESS AXIS NORMAL TO BAR AXIS

78.1 % REDUCTION IN THICKNESS AT 300° C.

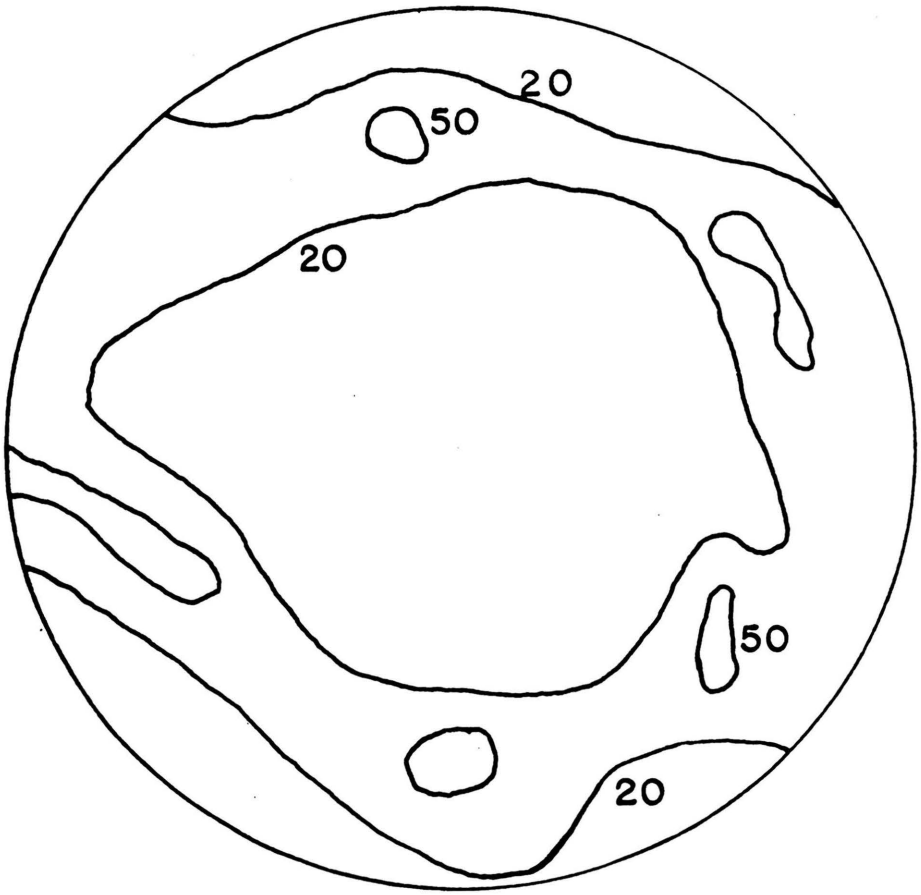


FIG. NO. 9

INNER 60° PORTION OF

(0002) POLE FIGURE

HAFNIUM CRYSTAL BAR COMPRESSION SAMPLE

STRESS AXIS PARALLEL TO BAR AXIS

87.2% REDUCTION IN THICKNESS AT 300° C.

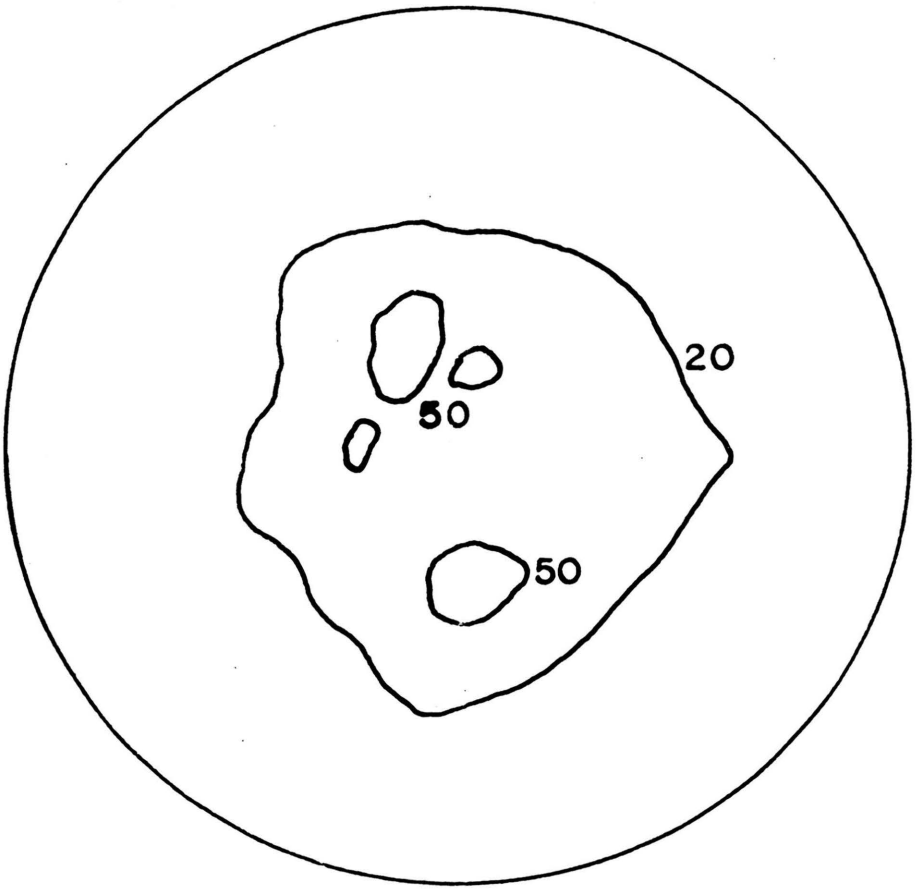


FIG. NO. 10

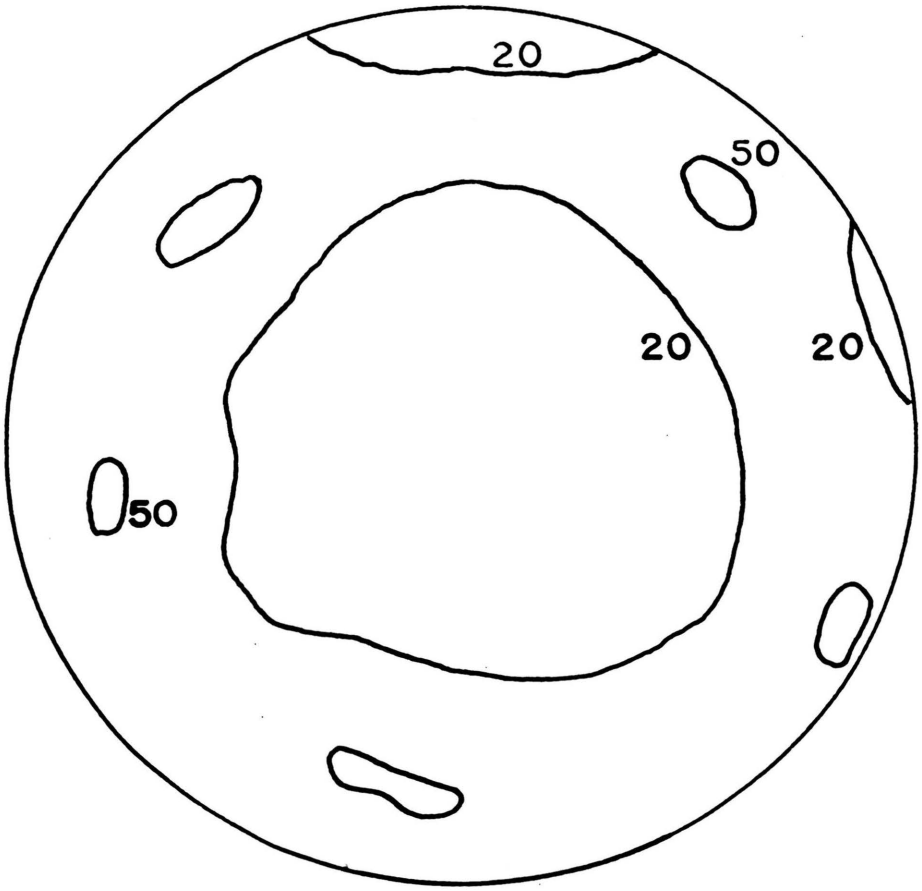
INNER 60° PORTION OF

$(10\bar{1}1)$ POLE FIGURE

HAFNIUM CRYSTAL BAR COMPRESSION SAMPLE

STRESS AXIS PARALLEL TO BAR AXIS

87.2 % REDUCTION IN THICKNESS AT 300° C.



rotation was 20 revolutions per minute while the drive was set for 1° per minute. The plots of two samples so run are shown on the following pages. The dotted line indicates the experimental intensity recorded and the solid line the intensity as corrected for tilting correction. For the (0002) plane the sum of these two graphs was the basis for the composite compression pole figure shown in Figure No. 13. This plot indicates a pronounced predominance of (0002) poles in the orbit between ϕ equals 20° and ϕ of 26°. On the basis of this determination the average angle of basal rotation can be said to be 23°.

This determination would tend to indicate that the compression texture for hafnium crystal bar material compressed at 300°C can be described as a (0002) plane rotated 23° out of the compression plane around an [10$\bar{1}$0] rotational axis.

D. WARM ROLLED TEXTURE

In that cold reduction was not possible to the extent necessary to develop any appreciable preferred orientation, it was decided to try to determine the warm rolled texture.

It is generally conceded that warm rolling will promote plasticity. It might be that this increase in plasticity is due to increased interplaner spacing of the operating slip system and therefore decreased resolved shear stress to bring about deformation. This explanation, therefore, allows increased plasticity without necessarily requiring any additional operating deformation mechanism. As long as the same slip and twinning mechanisms are assumed to be operating in warm rolling as in cold rolling, the resulting textures should be more

FIG. NO. 11

Ø ANGLE VS INTENSITY

CONTINUOUS α ROTATION OF CRYSTAL BAR COMPRESSION SAMPLES

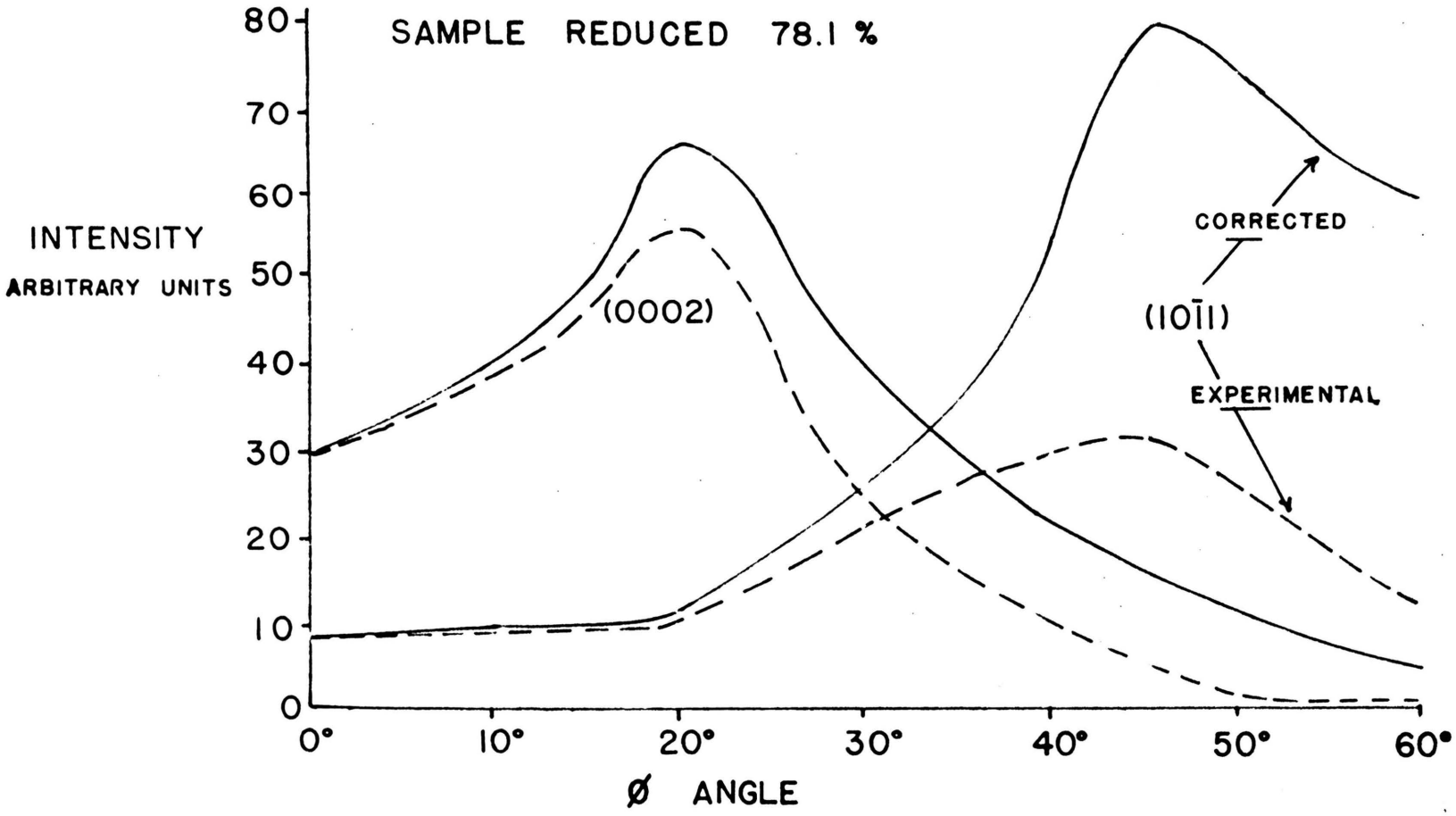


FIG. NO. 12

Ø ANGLE VS INTENSITY

CONTINUOUS α ROTATION OF CRYSTAL BAR COMPRESSION SAMPLE

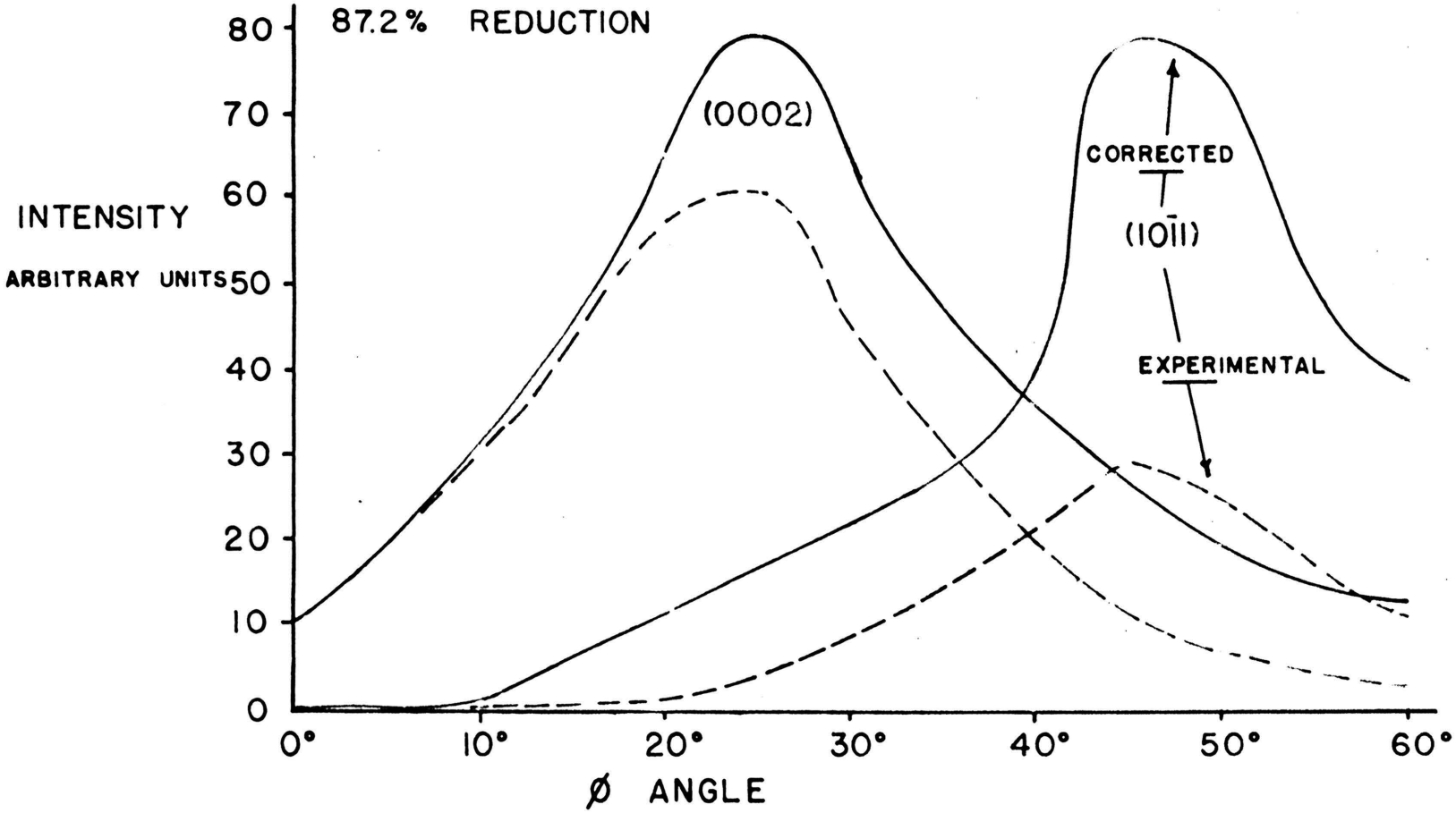
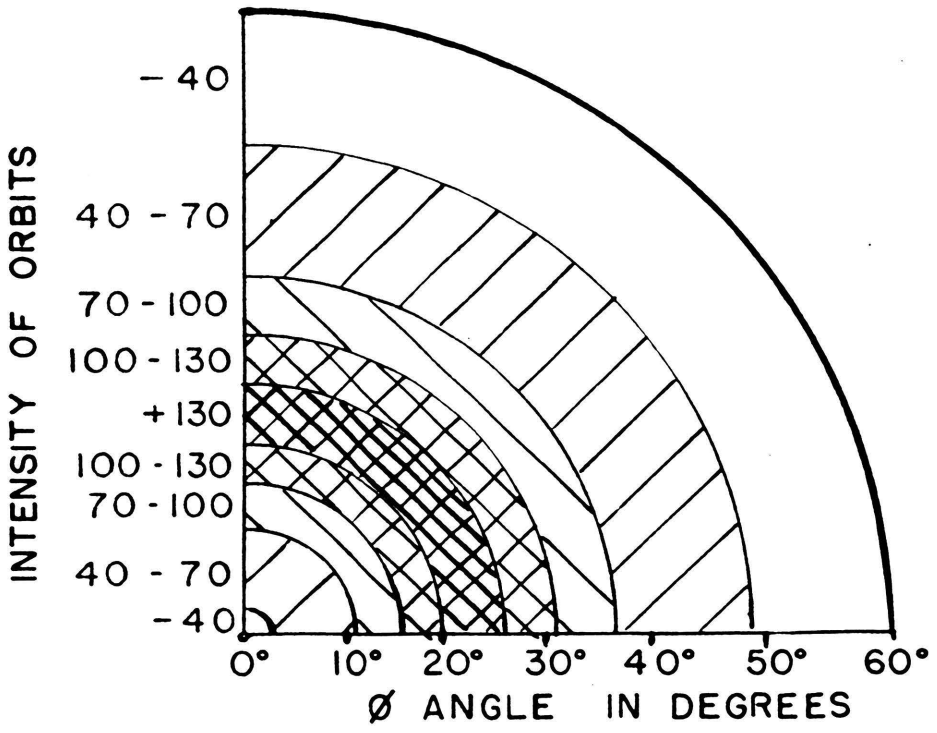


FIG. NO. 13

COMPOSITE (0002) COMPRESSION POLE FIGURE

DIAGRAM BELOW OBTAINED BY ADDING THE 87.2 % AND 78.1 % CORRECTED α ROTATED COMPRESSION FIGURES.

MAXIMUM INTENSITY AREA : 20° TO 26° ϕ ROTATION



ϕ EQUALS ANGLE BETWEEN COMPRESSION PLANE AND (0002)

or less similar.

It was decided to warm roll the crystal bar samples at 500°C, this temperature being several hundred degrees below the reported recrystallization temperature. The samples to be rolled, sectioned from the periphery of the crystal bar, were heated in a neutral salt bath to 500°C. The rolls were preset before the first pass and lowered approximately 0.002" to 0.003" between each pass. The sample was reheated approximately one minute in the salt bath between each pass. There was no scale formed on the sample during this repeated heating and rolling although the samples all became coated with a bluish coloration very similar in appearance to a temper color noted on steel. Although the samples were not noticeable oxidized, the rolled surface was very rough as a result of salt particles adhering to the sample and being rolled in. This surface roughness limited the ultimate reduction to approximately 0.020" thick samples. There was no noticeable cracking and it was felt that much greater total reductions would have been possible had thicker original pieces been available. After rolling the samples were processed on a facing grinder to obtain smooth flat surfaces. The resulting samples were approximately 0.010" thick.

The samples were then etched and the pole figures made in the conventional manner according to the description in the appendix. A representative series of the (0002), $(10\bar{1}1)$ and $(10\bar{1}0)$ pole figures is shown on the following pages.

It can be observed in the foregoing figures that the warm

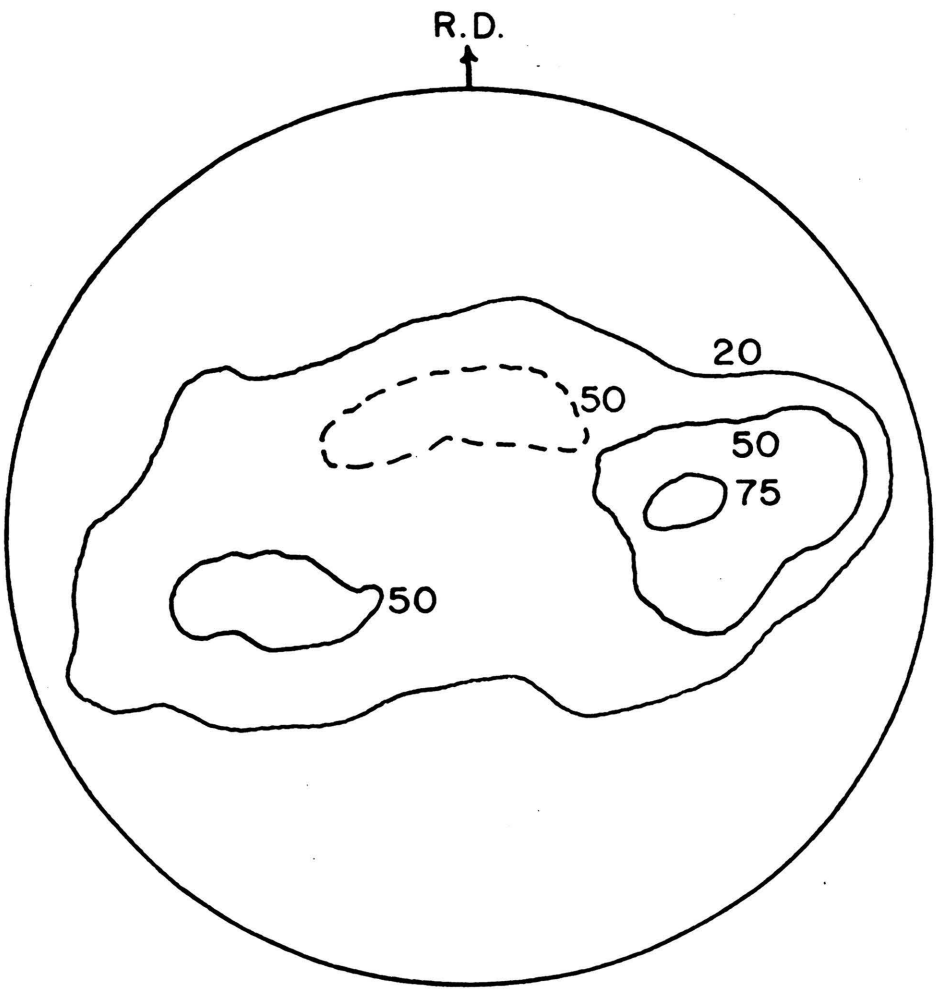
FIG. NO. 14

INNER 60° PORTION OF

(0002) POLE FIGURE

HAFNIUM CRYSTAL BAR WARM ROLLED SAMPLE

71.% REDUCTION IN THICKNESS AT 500° C.



DOTTED LINES INDICATE SPURIOUS AREA

FIG. NO. 15

INNER 60° PORTION OF

$(10\bar{1}1)$ POLE FIGURE

HAFNIUM CRYSTAL BAR WARM ROLLED SAMPLE

71. % REDUCTION IN THICKNESS AT 500° C.

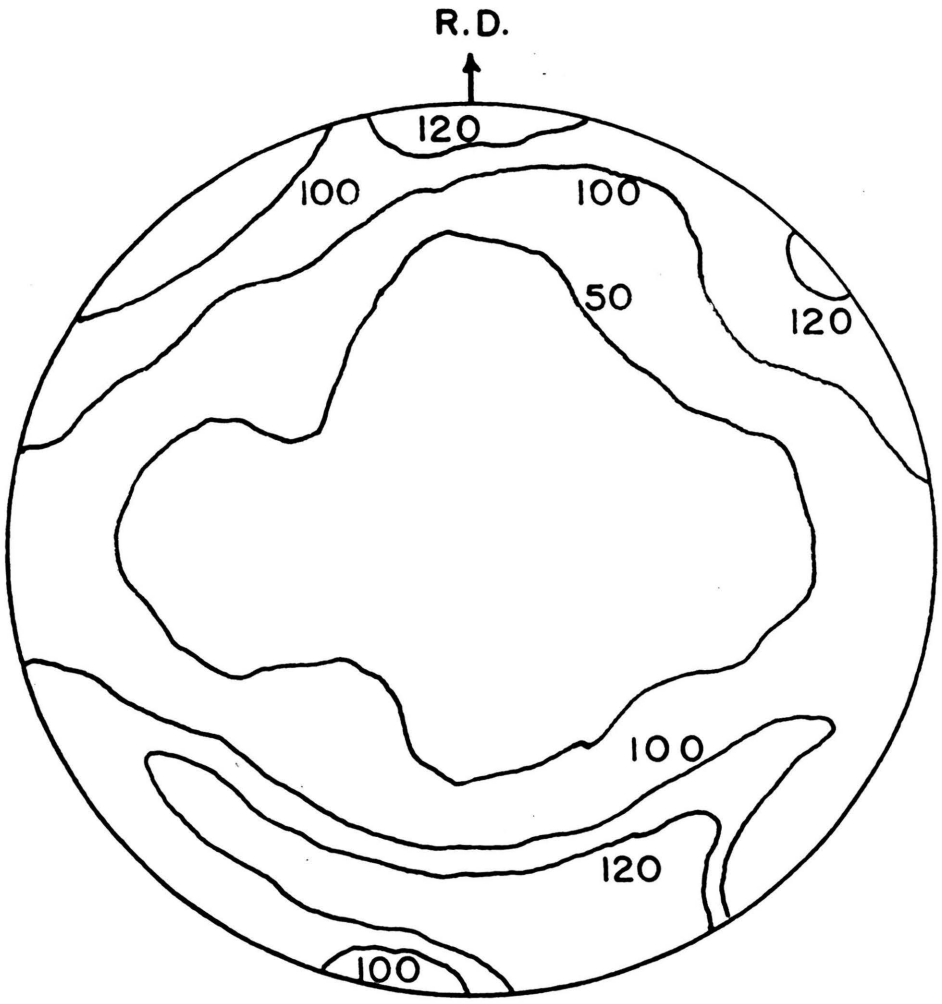


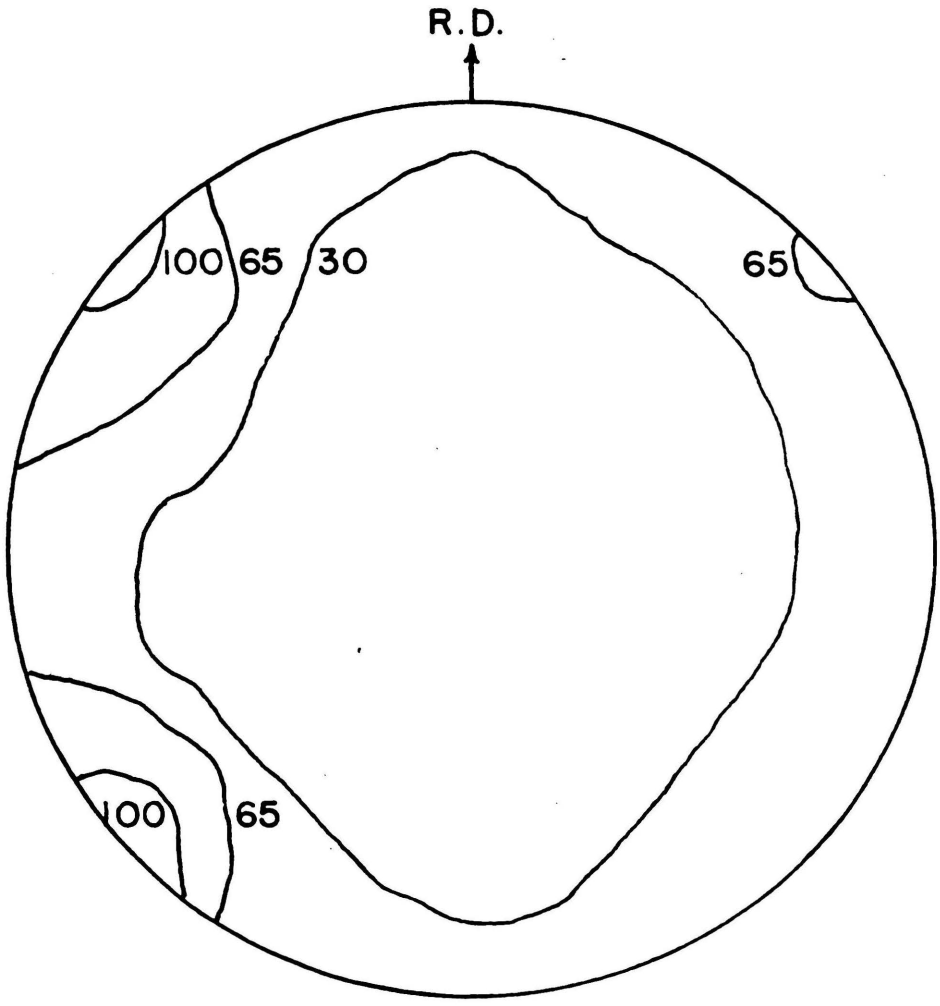
FIG. NO. 16

INNER 60° PORTION OF

(10$\bar{1}$0) POLE FIGURE

HAFNIUM CRYSTAL BAR WARM ROLLED SAMPLE

71.% REDUCTION IN THICKNESS AT 500° C.



rolled crystal bar material exhibits a (0001) $[10\bar{1}0]$ texture rotated 30° in the transverse direction.

This increase in angle of rotation over the cold rolled material reported in the next chapter may or may not be significant. If $\{10\bar{1}0\}\langle 11\bar{2}0\rangle$ and $\{10\bar{1}1\}\langle 11\bar{2}0\rangle$ slip and $(10\bar{1}2)$ and $(11\bar{2}2)$ twinning were accentuated at the expense of $\{0001\}\langle 11\bar{2}0\rangle$ slip such an increase in angle of transverse rotation would be expected. It may be that these mechanisms may be preferentially activated at warm rolling temperatures. This possibility is further supported by the very pronounced increase in width of the warm rolled samples. While cold rolling produces normally only a small increase in width, the warm rolled samples were approximately 50% wider after warm rolling. This too could be accounted for on the basis of increased prismatic and pyramidal slip at the expense on basal slip.

The (0001) $[10\bar{1}0]$ texture rotated 30° in the transverse direction is substantially similar to the other hexagonal metals in this class and to the cold rolled texture of hafnium subsequently ascertained.

CHAPTER IV

THE COLD ROLLED TEXTURE OF HAFNIUM

Subsequent to the work on samples sectioned from crystal bar, arc-melted material was obtained from the Bureau of Mines. These samples were of crystal bar which had been arc-melted and partially cold reduced. The chemistry was reported as being the same as the crystal bar samples previously supplied, Table No. 1.

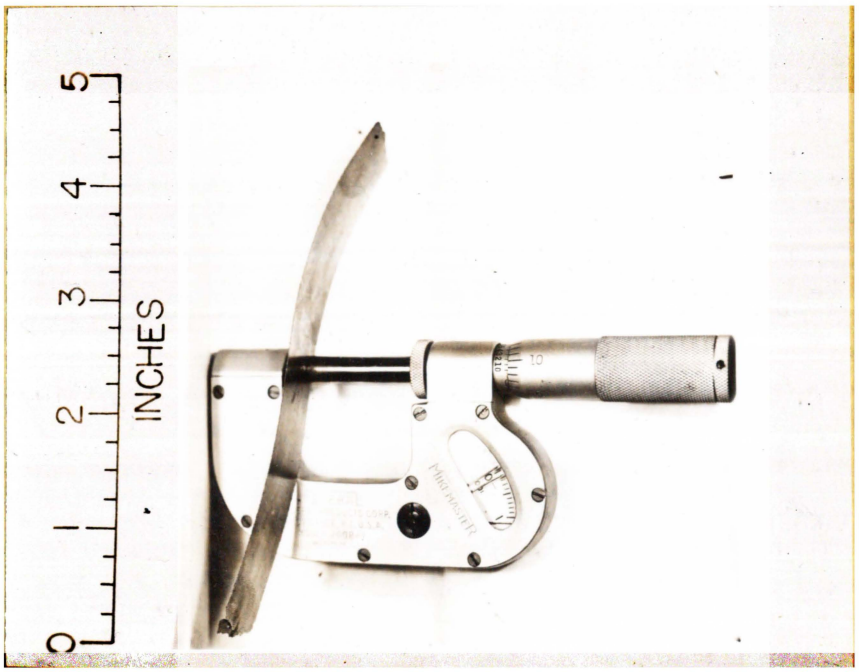
Using small reduction passes, samples of this material were cold rolled to the limit of the 2" rolls available. Each reduction pass was approximately 0.001" until the limit of the mill was approached where upon the reduction per pass decreased to approximately 0.0001" or less. Strips as thin as 0.004" were obtained. The samples were turned end-for-end after each pass. Only small amounts of edge cracking were evident in the final strip and it was found that this could be almost eliminated by grinding smooth rounded edges on the original samples. A typical cold rolled strip is shown in Figure 17.

A. HARDNESS OF COLD ROLLED STRIP

The hardness of the samples was checked in an annealed condition, at 50% reduction and at the ultimate reduction which in most cases was close to 95%. Preliminary readings indicated the difficulty of obtaining reproducible hardness values. A Tukon hardness tester with a diamond pyramid indentor was used. Because the samples were so thin, a 1 kilogram load was used. This was necessary to keep from penetrating the sample. Due to the relatively hard material, the re-

FIG. NO. 17

COLD ROLLED HAFNIUM STRIP



sulting indentations had a diagonal dimension of the order of 0.06 mm. Because the samples were so thin, it was impossible to do any mechanical polishing and as a result the accuracy of the diagonal measurement was no more than 0.001 mm. The formula for diamond pyramid hardness is:

$$D.P.H. = 1.854 \frac{P}{D^2}$$

where P equals load in kilograms and D is indentation diagonal in millimeters. It can be noted that a variation of $\pm$ .001 mm at a hardness level giving a diagonal indentation value of 0.06 mm will result in a variation of 17 D.P.H. points. Therefore it was assumed that the variation in readings might be as much as $\pm$ 20 points. To overcome this poor precision as much as possible, all determinations were repeated by a second individual on the same machine and rerun a third time by a third person using a second machine at the U. S. Bureau of Mines at Rolla, Mo. A plot of hardness versus percent reduction is given on the following page.

The procedure of averaging the hardness values is in part validated by a recent article on the accuracy of hardness readings by E. Rossow (44). His conclusion based on a statistical treatment of careful experimental data is that the average variation of Vickers' hardness values obtained by a single observer is 4%. He indicates this can be cut to 0.5 to 2% when determinations are repeated by

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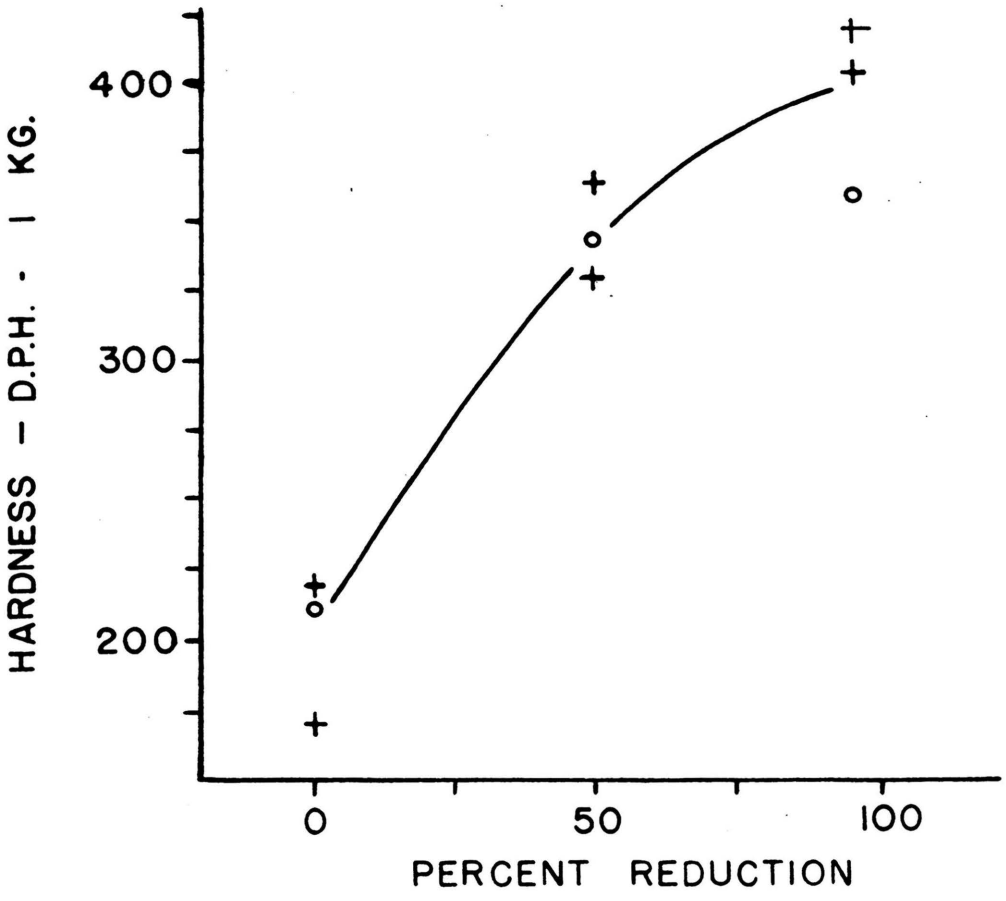
(44) Ernest Rossow, "Study of the accuracy of Vickers' and Rockwell hardness tests," Metaux, Corrosion, Ind., Vol. 30, pp. 476-483, 1955.

FIG. NO. 18

HARDNESS VS DEGREE COLD REDUCTION



\+ = M.S.M. HARDNESS TESTER

o = U.S.B.M. HARDNESS TESTER

several observations. A 4% variation at a hardness level of D.P.H. 400 would be 16 points.

An interesting point is that these readings are a great deal higher than zirconium. The average readings from Figure 18 are plotted with other data for hafnium and zirconium in Figure 19 (45, 46) The top curve denotes the experimental data from this investigation. It can be also noted from this graph that hafnium also appears to have a greater tendency to strain harden than does zirconium as can be noticed by the steeper slope of the hafnium curves.

B. REFLECTION DETERMINATION OF TEXTURE

The surfaces of the cold rolled strip were lightly polished with 4/0 grit paper and approximately 0.001" was removed by chemical etching from the surface to be examined. Preferred orientation determinations using the technique outlined in the appendix were then made. Pole figures for the (0002), $(10\bar{1}0)$ and $(10\bar{1}1)$ planes were made for a number of samples from different strips. A representative set from the same sample is shown by Figures 20, 21 and 22 on the following three pages.

The areas in the (0002) pole figures shown in dotted lines are actually spurious peaks. Such peaks in essentially the same places

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(45) Litton, Loc. cit.

(46) R. M. Treco, "Recrystallization and grain growth in iodide zirconium," J. of Metals, AIME, Vol. 8, p. 1305, (Oct.), 1956.

FIG. NO. 19

HARDNESS VS DEGREE COLD REDUCTION

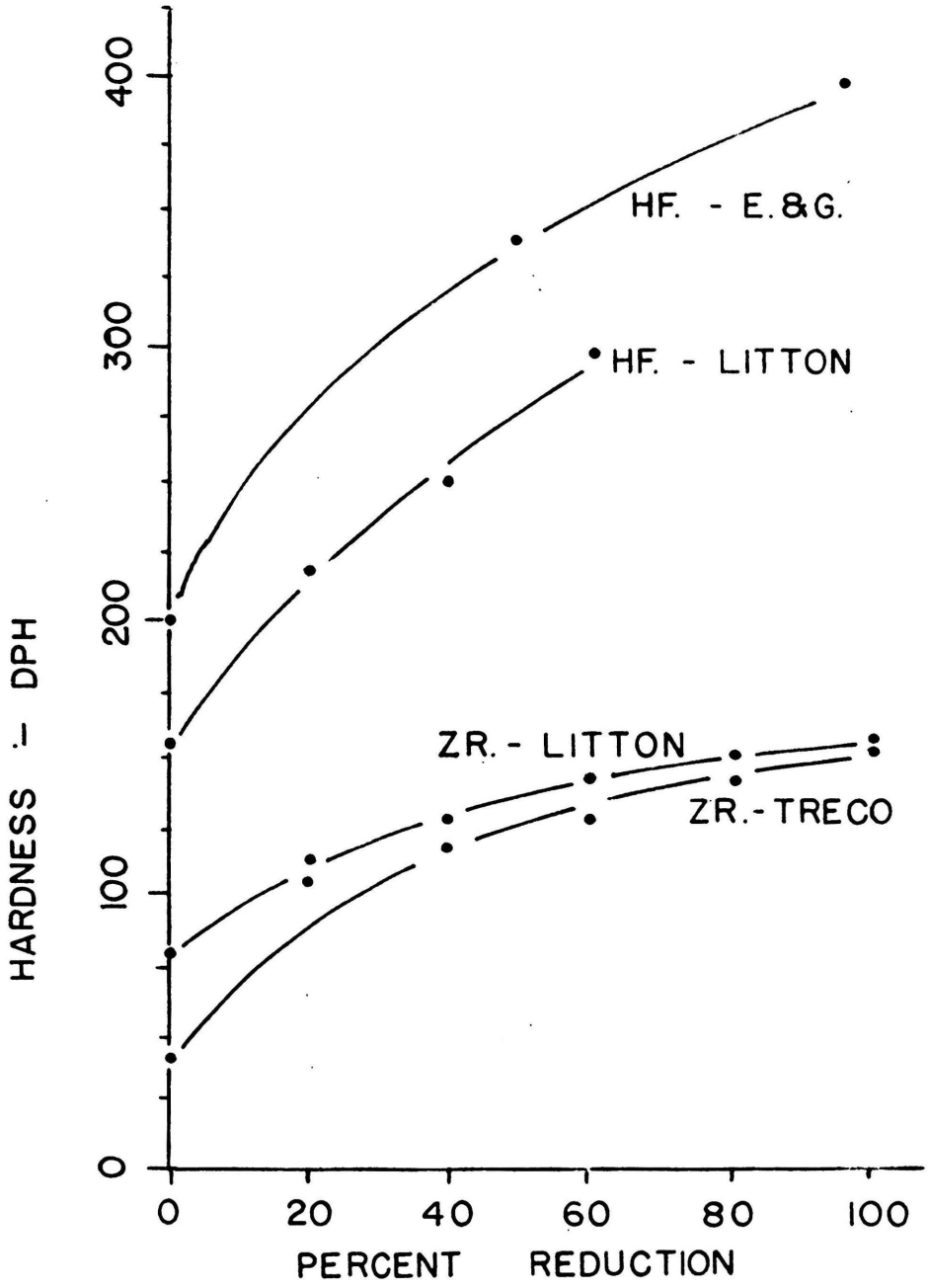


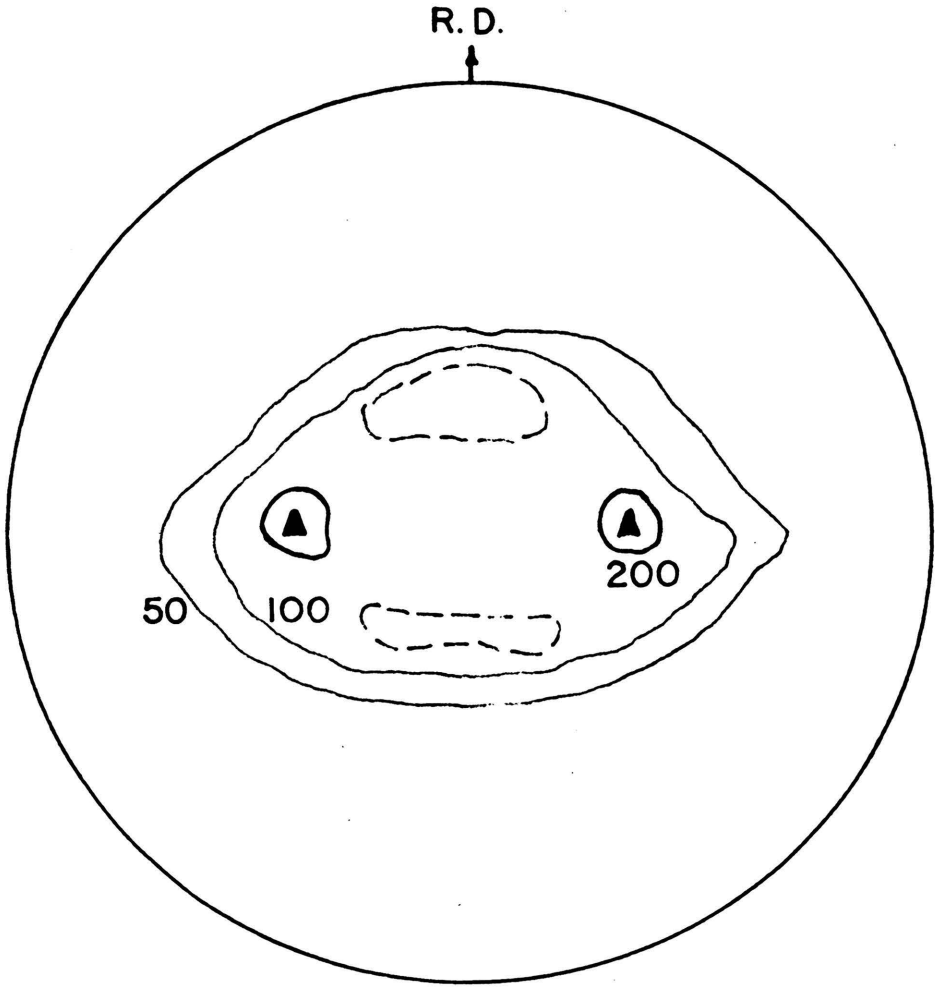
FIG. NO. 20

(0002) POLE FIGURE

HAFNIUM STRIP 95% COLD REDUCTION

INNER 60° PORTION ONLY

TRIANGLES INDICATE IDEAL (0001)[10$\bar{1}$0] TEXTURE
ROTATED 25° IN TRANSVERSE DIRECTION



DOTTED LINES INDICATE SPURIOUS AREAS

FIG. NO. 21

INNER 60° PORTION OF

$(10\bar{1}1)$ POLE FIGURE

HAFNIUM STRIP 95% COLD REDUCED

AS ROLLED - NO ANNEAL

TRIANGLES INDICATE IDEAL $(0001)[10\bar{1}0]$ TEXTURE

ROTATED 25° IN TRANSVERSE DIRECTION

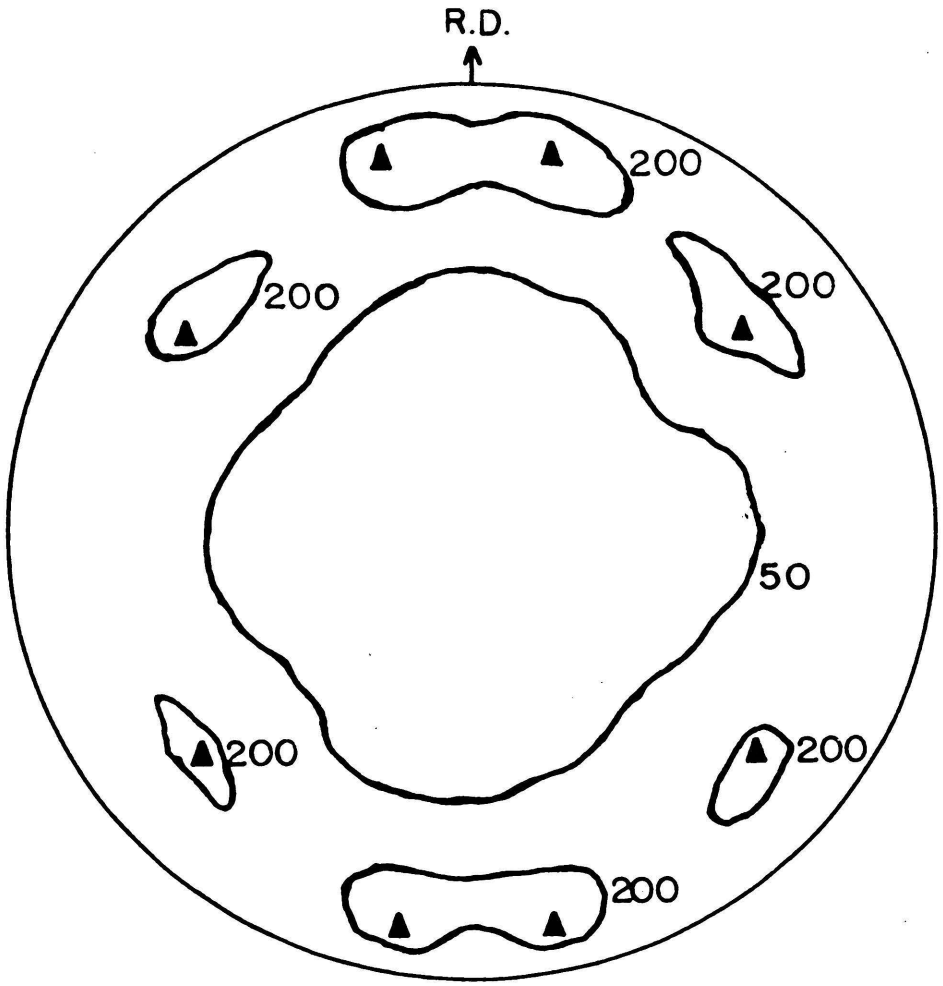
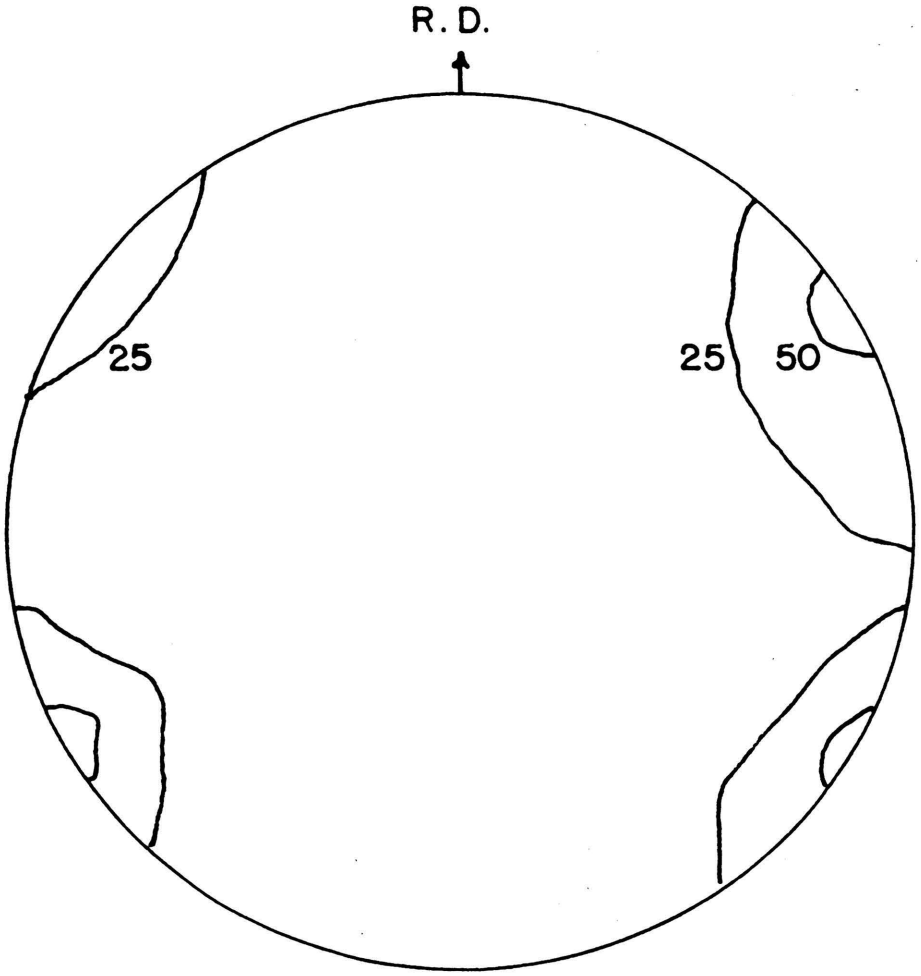


FIG. NO. 22

INNER 60° PORTION OF

(10$\bar{1}$0) POLE FIGURE

HAFNIUM STRIP 95% COLD REDUCTION



have also been observed by Geisler in titanium (0002) pole figures[47] These poles correspond generally to the positions of the $(10\bar{1}1)$ texture and are due to the lack of resolution of the diffraction. If they were true peaks, there would be double texture with a $\langle 10\bar{1}0 \rangle$ texture in the transverse direction. Examination of the $(10\bar{1}0)$ and $(10\bar{1}1)$ pole figures reveals that this is definitely not the case.

The (0002) and $(10\bar{1}1)$ poles occur at 2θ angles only one and one-half degrees apart. In order to try to resolve the $(10\bar{1}1)$ peaks out of the (0002) pole figures, 1/2° and 1/4° tube outlet slits were tried in place of the 1° slit otherwise used. These finer slits increased resolution, but at the expense of overall intensity. As a result of the decreased intensity the pole figures obtained with them were much less sharp. In that the lack of resolution of the 1° slit caused trouble only in the (0002) pole figure and since the nature of the spurious peaks was understood, the 1/2° and 1/4° slits were not subsequently used.

The small black triangles in the (0002) and $(10\bar{1}1)$ pole figures denote a (0001) $[10\bar{1}0]$ texture rotated 25° in the transverse. Assuming a precision measure for the equipment and technique of ± 2°, the cold rolled texture can then be described as a (0001) $\langle 10\bar{1}0 \rangle$ texture rotated 25° ± 2° in the transverse direction. This texture is shown schematically in Figure No. 23.

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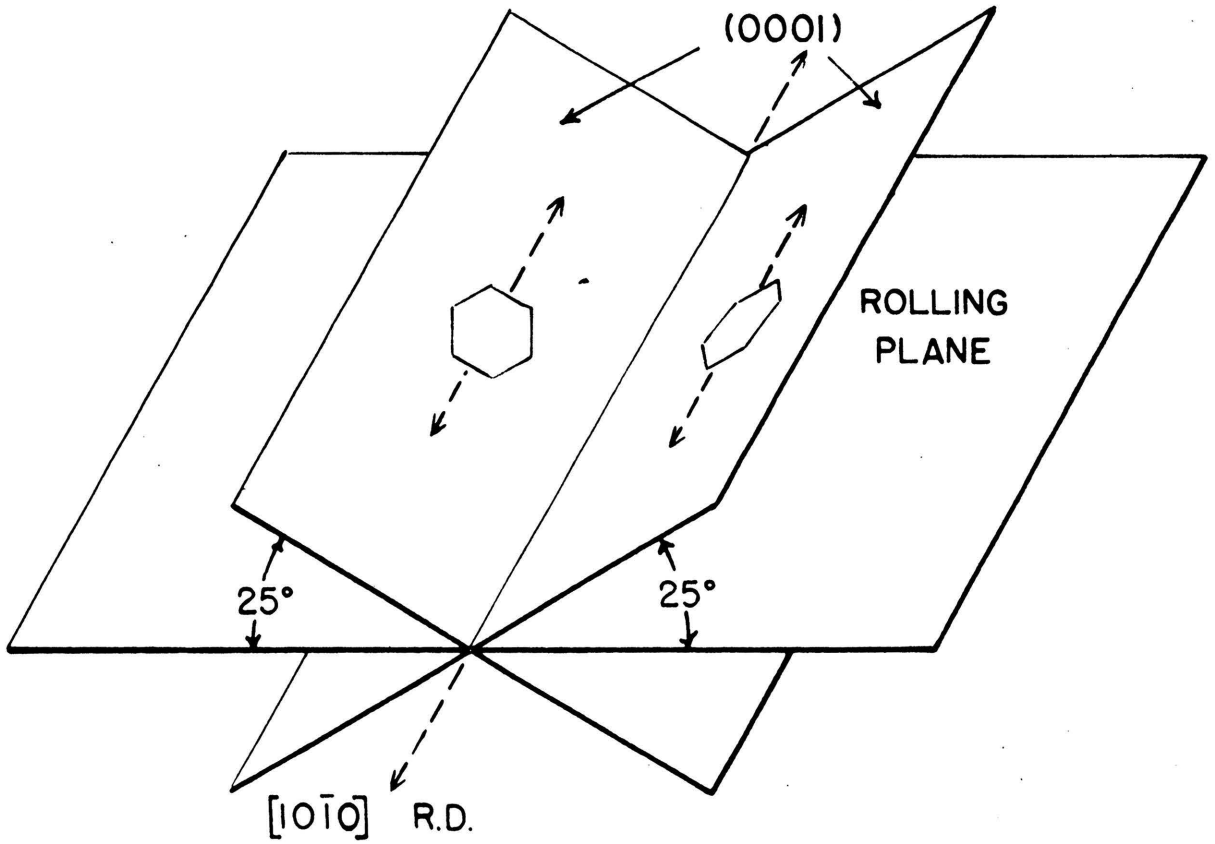
(47) Geisler, Loc. cit.

FIG. NO. 23

SCHEMATIC REPRESENTATION OF COLD ROLLED TEXTURE OF HAFNIUM STRIP



[10$\bar{1}$0] AND R.D. COINCIDENT

C. TRANSMISSION DETERMINATION OF PREFERRED ORIENTATION

Although the directionality of the cold rolled hafnium texture was evident on the basis of the reflection determinations, it is obvious that the $(10\bar{1}0)$ pole figure could have been more useful if the outer area had been run. In an effort to complete the whole pole figure an attempt was made to run the outer transmission area of the pole figure although transmission techniques have been generally unsuccessful for heavier metals[48].

A sample holder was made and mounted on the pole figure stage so that the sample was held in a position perpendicular to the reflection position. The sample holder was designed so that thin foils could be mounted for transmission diffraction. The motor drives of the fixture were reversed, the α rotation becoming the ϕ and the ϕ becoming the α. The actual mechanics of the method are treated in the appendix.

The problem with hafnium, relative to transmission diffraction, is its relatively high absorption of the CuK_α radiation. The mass coefficient of the absorption for hafnium amounts to only 157[49], which is not at all too high. The density of the material is, however, very high. The measured density, according to Litton[50], is 13.06

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(48) M. H. Mueller and H. W. Knott, "Quantitative pole figures for sheet material by the reflection technique," Rev. Sci. Inst., Vol. 25, p. 1115, 1954.

(49) H. P. Klug and E. E. Alexander, "X-Ray Diffraction Procedures," (New York: J. Wiley and Sons, 1954) p. 678.

(50) Litton, Loc. cit.

gms/cm^3. Geisler was able to get satisfactory transmission results from titanium foil of 0.001" thick[51]. In that titanium has approximately the same mass coefficient but only one third the density, it would appear that good transmission results could be obtained from hafnium 0.0003" thick.

Starting with the cold rolled strip 0.004 to 0.006" thick it was found impossible to etch the material to foil of average thickness less than 0.006" thick. This material, in spite of the most careful technique, was found to be uneven in thickness with holes etched all the way through. Etching beyond 0.0006" resulted in rapid increase in the size of the holes and edge corrosion resulting in complete destruction of the sample.

Small pieces of this 0.0006" thick foil were used for transmission preferred orientation studies. One quadrant of the $(10\bar{1}0)$ pole figure so obtained is shown in Figure 24 on the following page. Although this pole figure tends to verify the $[10\bar{1}0]$ directionality of the cold rolled material, it is generally unsatisfactory. The magnitude of the intensities recorded is very small and therefore more or less unreliable. The intensities away from the periphery up to 60° from the center of the pole figure were corrected according to Decker's absorption correction formula[52]. However, it was felt that the correction was not valid any further toward the center of the figure due

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(51) Geisler, Loc. cit.

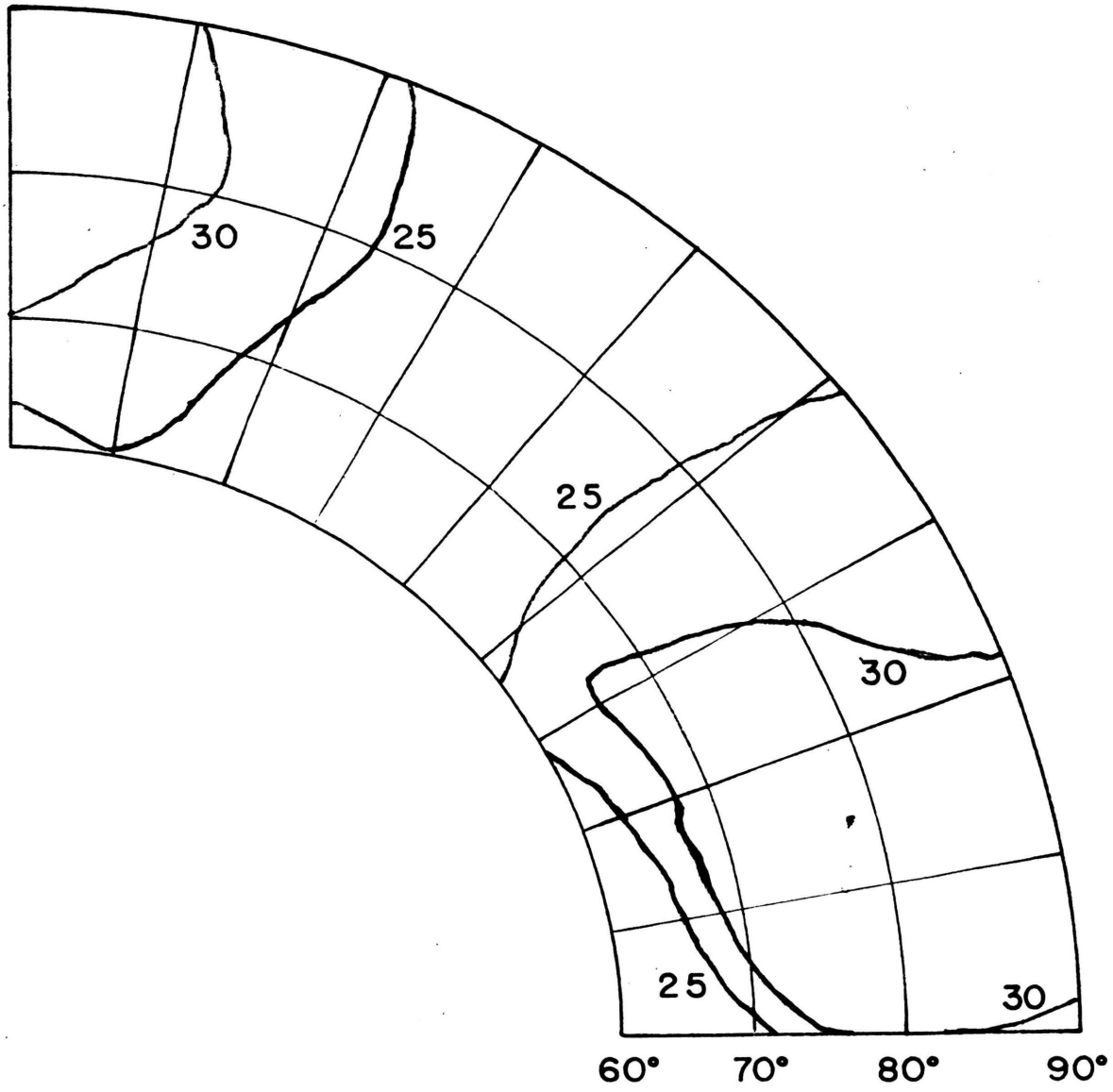
(52) B. F. Decker, E. T. Asp and D. Harker, "Preferred orientation determination using a Geiger counter X-ray diffraction goniometer," J. Appl. Phy., Vol. 19, p. 388.

FIG. NO. 24

(10$\bar{1}$0) POLE FIGURE

TRANSMISSION SEGMENT

HAFNIUM STRIP 95% COLD REDUCED



INTENSITY CONTOURS IN COUNTS PER SECOND;
INNER POLES CORRECTED FOR ABSORBTION

to the rather large influence of background in relation to the measured diffracted peak. For this reason only the outer thirty degrees were used and no attempt was made to correlate the transmission and reflection pole figures.

CHAPTER V

THEORETICAL DISCUSSION OF COLD ROLLED TEXTURES OF HAFNIUM AND OTHER HEXAGONAL METALS

Previous workers have definitely established the textures of a large number of cold rolled hexagonal metals. This study has resolved the cold rolled texture of hafnium. The purpose of this discussion is to account for the cold rolled texture of hafnium and to relate it to the already determined textures of zirconium and titanium. The discussion is further extended to attempt to relate the textures of these three metals to the whole hexagonal system on the basis c/a ratio.

A. ORIGIN OF TEXTURES IN HEXAGONAL METALS

For purposes of this discussion the hexagonal metals will be divided into three classes on the basis of c/a ratio. This classification is obvious on the basis of the twenty-one close packed hexagonal metals listed in the Metals Handbook[53]. Zinc and cadmium have c/a ratios of 1.86 and 1.89 respectively, thirteen metals have c/a ratios between 1.63 and 1.61 and six have c/a ratios of 1.60 or less. Of course the dividing line between the second and third groups is not as well defined as between the first two and as might be expected there is not as marked a change in textures as between the first two.

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(53) "Metals Handbook," 1948 Edition, (Cleveland, American Society for Metals, 1948), p. 21.

Previous work on the generalization of the development of textures (54) has divided the hexagonal system into two groups based on $c \pm \sqrt{3}a$. This of course put cadmium and zinc in one class and all the other metals in a second class. It has become evident, however, that as the c/a ratio drops much below ideal packing, 1.633, new deformation mechanisms become operative at the expense of the more common basal slip and simple pyramidal twinning.

In further laying the groundwork for this discussion, it is necessary to make a number of generalized assumptions; some very well established, some very specialized and applying only to this specific discussion.

In general it is believed that the shear stress necessary to make a particular slip system operative in a given metal is a function of interplaner spacing. Planes with a large interplaner spacing, and therefore high atomic density in the plane, have the lowest shear stress values. Such planes, therefore, of large spacing and high density are usually the preferred slip planes. The same general assumption also applies to twinning in that shearing force parallel to the twin plane with atomic movement in that direction also exists. As far as atomic movement is concerned twinning can most generally be considered as a homogeneous slipping, with atomic movement generally parallel to the twin plane. The specific slip and twin planes in the hexagonal systems will be con sidered later.

It is also further assumed that the direction of slip will al-

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(54) Calnan and Clews, Loc. cit., (Ref. 36).

ways be coincident with the most closely packed rows of atoms in the lattice. This requires rotation of the lattice plane to accommodate the stress axis. It is normally considered that the slip plane normal will rotate to the compression axis and the slip plane direction will rotate toward the tensile axis. As will be noted later such rotation can place the lattice in such a position that more complex slip or twinning can proceed.

The preceding deformation mechanisms are obvious for unrestrained single crystals. The deformation of polycrystalline materials is considerably more complicated. In the case of the single crystal the lattice is able to move without the restraint of the surrounding grains that exist in a polycrystalline material. As a result, in polycrystalline materials it is assumed that multiple deformation mechanisms occur simultaneously to avoid grain boundary separation. So therefore, slip and twinning in polycrystalline materials must provide for the overall change in shape of the polycrystalline mass and also maintain grain boundary cohesion. It will be seen that this requirement demands multiple slip and also suppresses some deformation mechanisms which can only proceed where no grain boundary restraints are imposed. This concept provides the assumption that in a polycrystalline material the effective stress on a particular lattice may not necessarily coincide exactly with the applied stress as a result of the stress applied to the lattice by surrounding grains.

Further in considering cold rolling, it is assumed that the deformation in the rolling direction is the result of tensile stress

and in the transverse direction the result of compressive stress. Therefore in cold rolling both tensile and compressive deformation mechanisms are active. In regard to this, it is logical to assume that only mechanisms which can bring about a reduction in thickness of the rolled material will be in operation. It will be noted later that this requirement will eliminate the possibility of some deformation mechanisms operating in spite of the fact that the resolved shear stress may exceed the critical value required.

Origin of textures in first class - zinc. As might well be expected with a large c/a ratio, the usual slip system in zinc is (0001). The basal plane slips in a $[11\bar{2}0]$ direction resulting in slip plane rotation so that the $[11\bar{2}0]$ direction coincides with the rolling direction. Basal slip also causes the (0001) plane to be rotated into the rolling plane as deformation continues. As the basal plane approaches the rolling plane $\{10\bar{1}2\}$ type twinning can and does take place causing the material to lengthen in the tensile direction and further reorient the basal plane so that more basal slip is possible. Twinning of the above type can and does take place in tension and compression causing the pole of the basal plane to spread over quite an angle away from the rolling direction, but at any rate the basal pole is reoriented at an angle of approximately 60° away from the rolling plane normal allowing further slip as noted above. It would appear the metals such as zinc would be quite ductile in that these two mechanisms, basal slip and $\{10\bar{1}2\}$ twinning, are complimentary; each rotating the lattice into a desirable position for

the other. Single crystals of zinc have been found to be very ductile[55]; however, cold rolled polycrystalline zinc is usually warm rolled at several hundred degrees to overcome the rather poor rolling characteristics of the material. The poor cold rollability of pure zinc can easily be accounted for on the basis of the foregoing deformation mechanism. With only compressive twinning accounting for deformation other than in the rolling direction and this accounting for only a small amount of deformation at the most, the material undoubtedly fails of the result of intergranular separation. If there were a more complex slip mechanism operating, zinc would undoubtedly be more ductile as the grains would be able to deform as a result of the stress applied on each other, thereby providing better intergranular cohesion.

It might be expected that zinc might not have a well defined terminal texture as a result of the continuous movement of the basal poles but normally the greatest concentration is found 20° to 25° from the rolling plane normal in the rolling direction[56]. The orientation is almost intermediate between the position of the basal plane and rolling plane after twinning and the position for twinning to take place.

Origin of texture in second class - magnesium. The cold rolled texture of magnesium has been well established for a number of

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(55) Schmid and Boas, Loc. cit.

(56) Barrett, Loc. Cit.

years[57]. It is essentially a (0001) texture, the basal plane in the rolling plane, with definite $[11\bar{2}0]$ directionality. A slight spread of the basal plane normals in the rolling direction is also observed. The definite spread of the basal poles along the rolling direction and completely out of the center of the pole figure, evident in zinc, is not observed. This (0001) $[11\bar{2}0]$ texture tends to indicate that in magnesium, as in zinc, basal slip is a major deformation mechanism.

In the crystal lattice of magnesium, because of the smaller c/a ratio, it can be seen that the $(10\bar{1}2)$ twin plane makes an angle greater than 45° with the hexagonal axis. As a result $\{10\bar{1}2\}$ twinning cannot occur when the hexagonal axis is normal to the rolling plane - the end position of (0001) slip in tension and compression. Because $\{10\bar{1}2\}$ twinning cannot operate without increasing the thickness of the material, a violation of a fundamental premise, the basal plane is not rotated into a position for secondary slip. As soon as all the grains of the cold rolled material are orientated with the basal plane in the rolling plane, failure is sure to occur upon further deformation. The experimentally determined texture of magnesium is shown in Figure 25.

There are other mechanisms, which, it would seem, could tend to place the basal planes in a position for further slip; but apparently as a result of the packing arrangement of the atoms in the particular lattice, none have critical shear stress values low enough to be

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(57) P. W. Bakarian, "The preferred orientation in rolled magnesium and alloys," Metals Tech., Tech. Pub., AIME, 1355, August, 1941.

activated. As a result pure magnesium is a very difficult material to cold roll.

It has been shown that in both of the foregoing cases, alloying elements can increase the amount of cold rolling possible and do effect the resulting texture. It is obvious the two phenomena are related. It would appear that any element that could change the c/a ratio in magnesium or could alter the critical shear stress values in either one could provide additional operating deformation mechanisms without necessarily producing a new phase in the microstructure. Any additional slip mechanism or twinning system would certainly improve cold rollability.

Origin of textures in the third class - titanium. The texture of titanium has been found to be a (0001) $[10\bar{1}0]$ texture rotated approximately 30° in the transverse direction[58]. The texture was determined experimentally before any work was reported on the deformation mechanism. The $[10\bar{1}0]$ directionality and basal pole rotation in the transverse direction denote a more complex mechanism of deformation than the simple basal slip and $\{10\bar{1}2\}$ twinning. Williams and Eppelsheimer rationalized the experimental texture on the basis of pyramidal slip, which had been postulated theoretically, plus prismatic slip and several types of twinning[59]. These deformation mechanisms were all subsequently found to exist and be operative. Either $(10\bar{1}1)$ or $(10\bar{1}0)$ slip in a $[10\bar{1}2]$ direction will cause $[10\bar{1}0]$

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(58) Williams and Eppelsheimer, Loc. cit., (Ref. 3).

(59) Williams and Eppelsheimer, Loc. cit., (Ref. 3).

directionality. Twinning of the type of $\{10\bar{1}1\}$, $\{11\bar{2}1\}$, or $\{11\bar{2}2\}$ is required to remove the basal poles away from the rolling plane normal. By making assumptions as to the ratios of the critical shear stress values of the assumed systems along with application of the Calnan and Clews technique[60] Williams and Eppelsheimer were able to show the theoretical basis for the experimentally determined texture of titanium[61]. On the basis of the relatively larger number of operating deformation mechanisms, it might be assumed that the relatively poor rolling characteristics occurring in the first two group metals, would not occur in titanium. It can be postulated that greater total deformation is possible on metals in the third group and this assumption has been more or less experimentally verified.

It has been shown, however, that almost all impurities in titanium tend to make it harder to deform the material[62]. This is most probably due to the effect of impurities in blocking some of the operating deformation mechanisms. If any deformation system were blocked it would, of course, alter the resulting texture. This, in the case of aluminum in titanium, has been found to be the condition which exists[63].

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(60) Calnan and Clews, Loc. cit.

(61) Williams and Eppelsheimer, op. cit.

(62) T. D. McKinley, "The effect of impurities on the hardness of titanium," J. of the Electrochemical Society, October, 1956, p. 561.

(63) C. J. Sparks, Jr., C. J. McHargue, and J. P. Hammond, "Effects of aluminum on the cold-rolled textures of titanium," J. of Metals, AIME, January, 1957, p. 49.

B. APPLICATION OF THE CALNAN AND CLEWS TECHNIQUE OF TEXTURE ANALYSIS TO HEXAGONAL METALS

Briefly Calnan and Clews technique is a graphical method of texture analysis based on combining the rotational tendency of each slip and twinning tendency in a single crystal. Resolved shear stress contours are plotted in the stereographic unit triangle for each slip and twinning system known to be active. The resolved shear stress function plotted, in the case of slip, is product of the cosine of the angle of the stress axis and the slip plane normal and the cosine of the angle between the stress angle and the direction of slip. The lattice rotational tendencies of each system are then plotted on the basis of the resolved shear diagrams.

A unique feature of the system is the concept that the effective stress is free to rotate away from the axis of applied stress. This can theoretically keep slip from occurring at points of highest resolved shear stress and allow it to proceed at lower values of resolved shear stress where in some cases duplex slip can occur. Duplex slip is assumed to operate when the actuating resolved shear stresses are equal. This concept is one of the superior features of the Calnan and Clews technique.

After the slip and twinning diagrams in tension and compression are obtained they are combined to form an overall diagram which indicates the end positions. These end positions can be adjusted if the ratio of critical shear stress of the different operating systems are known and precise theoretical pole figures can be thus obtained.

For a more complete explanation of the technique other than the original paper[64], the reader is referred to the doctoral thesis of D. N. Williams[65]

In the following illustration, Calnan and Clews theoretical texture diagrams for generalizing the three hexagonal metal classes are given. These are essentially the same as the experimentally determined cold rolled textures plotted along side them.

C. APPLICATION OF CALNAN AND CLEWS TECHNIQUE TO THE THEORETICAL DEFORMATION TEXTURE OF HAFNIUM

Previous study of titanium. As a part of his study of the deformation texture of titanium, Williams[66] applied the Calnan and Clews method in a theoretical study of the experimentally determined texture of titanium. His only absolute data were the experimentally determined compression, tensile and cold rolled textures of titanium. At this time, 1953, the data as to the operating deformation mechanisms was incomplete. Williams was therefore forced to assume that the systems which were subsequently found to actually exist did, in fact, operate. At this time basal slip had not been observed, but it is necessary, on the basis of the Calnan and Clews method, to assume it did exist in order to explain the experimentally determined structure. The fact that it was subsequently discovered is a substantial valida-

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(64) Calnan and Clews, Loc. cit., (Ref. 36)

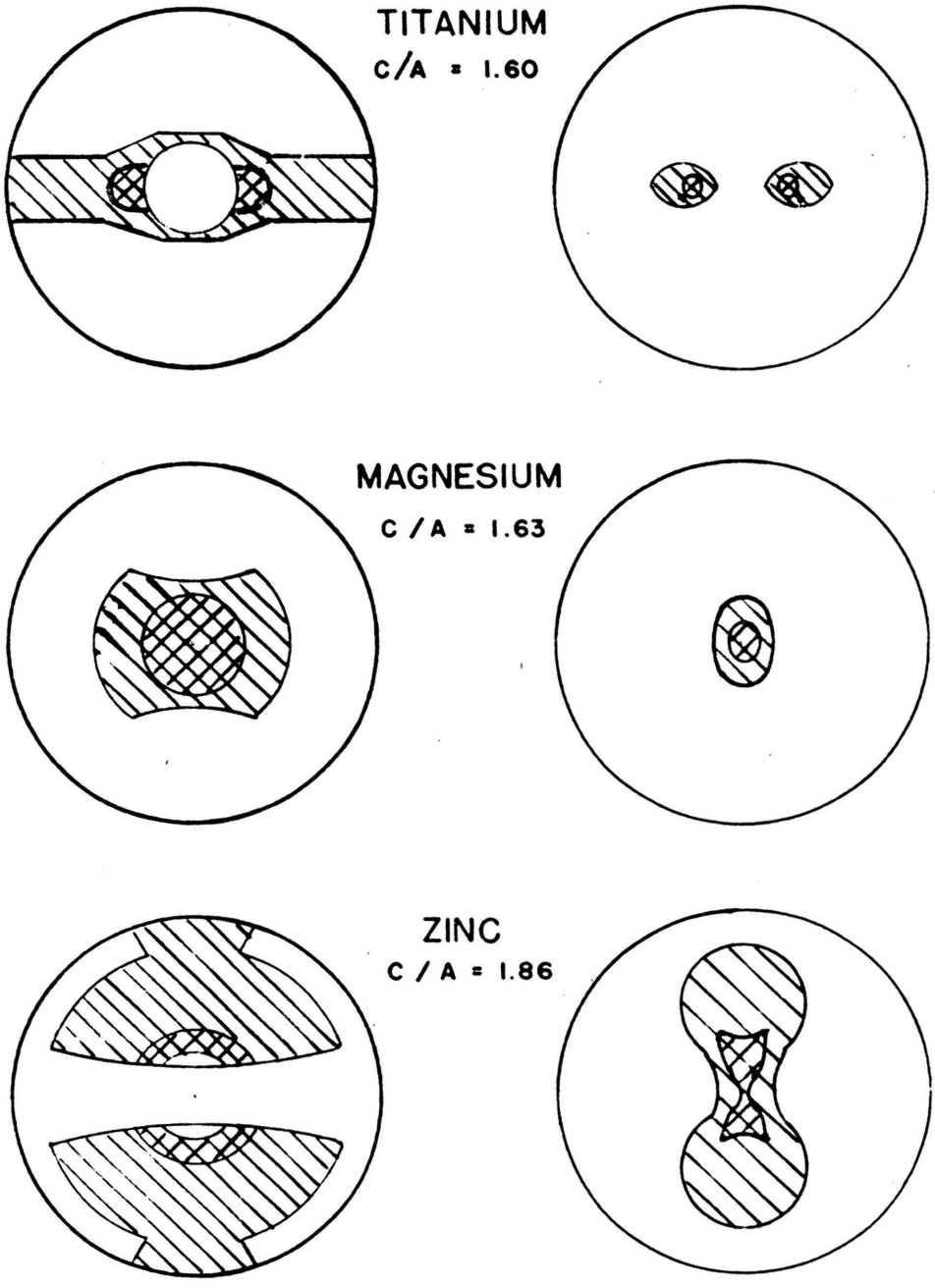
(65) Williams, Loc. cit., (Ref. 4).

(66) Williams, Loc. cit.

FIG. NO. 25

THEORETICAL VS EXPERIMENTAL TEXTURES
HEXAGONAL METALS

CALNEN & CLEWS ANALYSIS EXPERIMENTALLY DETERMINED



(0001) POLE FIGURES

tion of the method. The systems which have been reported are tabulated on the following page.

All of this information relative to the operating deformation mechanisms was obtained from the distortion of single crystals. This in part would tend to explain why all the possible mechanisms were not found by each investigator working with a limited number of samples. A randomly oriented material would be more apt to exhibit all or most all of the possible mechanisms as a result of every possible lattice orientation being subjected the shearing stress. In a single crystal some of the systems are bound to be initially oriented so that the resolved shear is too low to allow movement.

Anderson, Jillson and Dunbar[67] also postulate that the grain size of the single crystals studied may have some effect on the slip systems observed. They suggest that basal slip might not occur in very fine grains.

Another reason for the predominance of prismatic slip at the expense of basal slip is the large difference in critical shear stress for the systems. A value of 10,000 to 13,000 gms./mm^2 is indicated for (0002) $[\bar{1}120]$ slip, while (1010) $[\bar{1}120]$ slip appears to have a critical shear stress of approximately 5,000 to 6,000 gms./mm^2[68]. This variation will be discussed later in relation to Williams evalua-

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(67) Anderson, Jillson and Dunbar, Loc. cit.

(68) F. D. Rosi, "Mechanism of Plastic Flow in Titanium: manifestation and dynamics of glide," J. of Metals, AIME, p. 58, (Jan.), 1954.

TABLE II

IDENTIFICATION OF DEFORMATION PLANES IN TITANIUM

Mechanism	Plane	Observer (1)	(2)	(3)
Slip	$(10\bar{1}0)$	X	(a)	X
Slip	$(10\bar{1}1)$	X	(a)	(b)
Slip	(0002)	(b)	(a)	X
Twinning	$(10\bar{1}2)$	X	X	X
Twinning	$(11\bar{2}1)$	X	X	X
Twinning	$(11\bar{2}2)$	X	X	X
Twinning	$(11\bar{2}3)$	(b)	X	(b)
Twinning	$(11\bar{2}4)$	(b)	X	(b)

(a) Not reported - apparently not looked for

(b) Did not find

Observer (1) = Rosi, Dubi and Alexander, Loc. cit.

Observer (2) = Liu, Steinberg, Loc. cit.

Observer (3) = Anderson, Jillson and Dunbar, Loc. cit.

tion of the ratio of the critical shear stresses of the operating systems based on theoretical assumptions.

Although there has been no work reported on the operating deformation mechanisms for hafnium, it is logical to assume that the same systems operating in titanium will also exist in hafnium. This conclusion is predicted on two facts. First, the deformation mechanisms operating in titanium were predicted of the basis of crystallographic arrangement (69). In that titanium, zirconium and hafnium all have basically the same crystallographic arrangement, the same systems can be predicted in each. This is further substantiated by the fact that variable, although small, amounts of one of the above metals in another, i.e. Zr in Ti, Hf in Zr, Zr in Ti etc. seem to have little or no effect on deformation characteristics of the base material. This would seem to indicate similar mechanisms in each. The second substantiating fact is that the experimentally determined textures for all three are strikingly similar. This can only be rationalized by granting that similar deformation processes were followed. It is not necessary to know or assume the exact values for the critical shear stress forces, but it is logical to assume that the relationship between them is approximately the same for all three metals. As long as the terminal texture is the same for all three of the metals, it is fair to assume that the deformation mechanisms by which these textures were obtained were essentially the same whether or not the force necessary to obtain these deformations was the same

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(69) Rosi, Loc. cit.

or not.

As a result of the fact that hafnium has the same crystallographic arrangement as titanium and has essentially the same terminal texture after an equivalent amount of deformation, it would seem to be possible to assume that the same slip and twinning systems found for titanium would also be present in hafnium. If the foregoing is allowed, then the Calnan and Clews method for texture analysis can be applied to hafnium in a manner similar to that used for titanium by Williams and Eppelsheimer[70].

Lattice rotation and texture resulting from slip. On the following pages the tension and compression slip rotations resulting from (1011) $[\bar{1}120]$, (0001) $[\bar{1}120]$ and (1010) $[\bar{1}120]$ slip are shown. The (1010) $[\bar{1}120]$ and (0001) $[\bar{1}120]$ diagrams are exactly the same as for titanium. The (1011) $[\bar{1}120]$ are slightly different as a result of the slight difference in c/a ratio.

These three diagrams are combined on the fourth page, Figure Number 29. Without any consideration of any one slip system being predominate as a result of variation of critical shear stress values, the basis slip rotation and texture can be rationalized. All three tension systems have a $[\bar{1}010]$ component and it is particularly strong in the (1010) $[\bar{1}120]$ and (1011) $[\bar{1}120]$ systems. This accounts for the $[\bar{1}010]$ directionality of the resulting cold rolled texture. The two predominate textures in the combined diagram are a $[0001]$ and $[\bar{1}120]$. The experimental texture indicates the preferred position is along the

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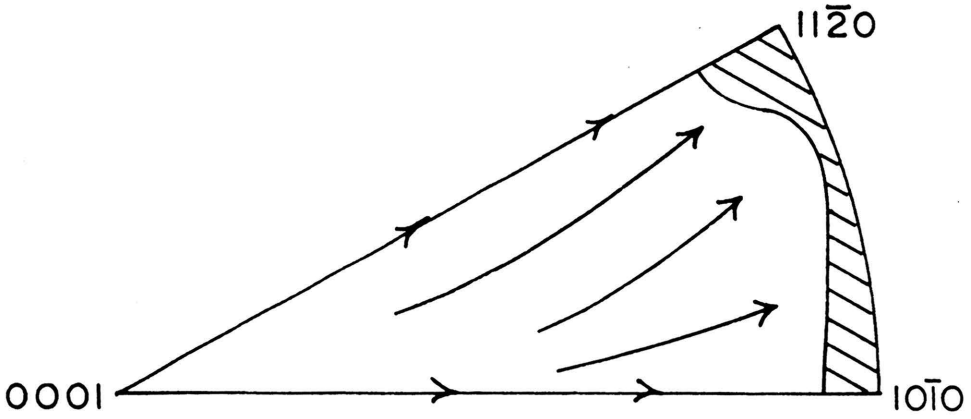
(70) Williams and Eppelsheimer, Loc. cit., (Ref. 3).

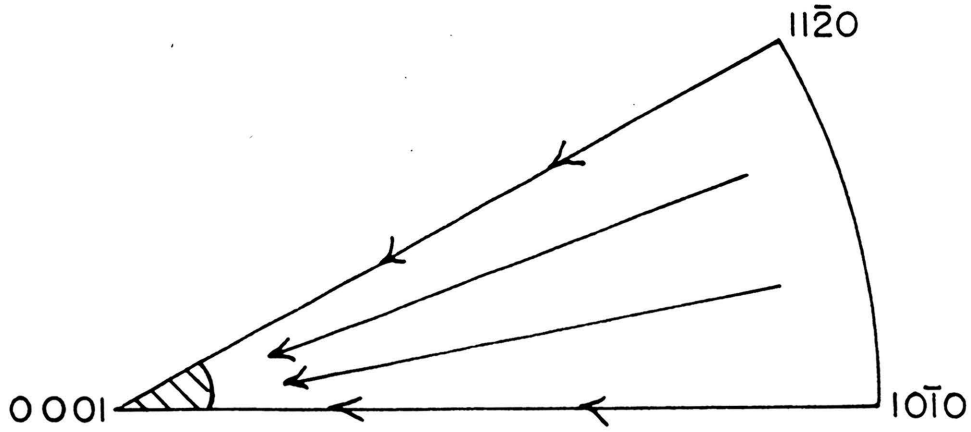
FIG. NO. 26

TENSION & COMPRESSION SLIP ROTATIONS AND TEXTURE FOR (0001) [11$\bar{2}$0] SLIP

TENSION



COMPRESSION

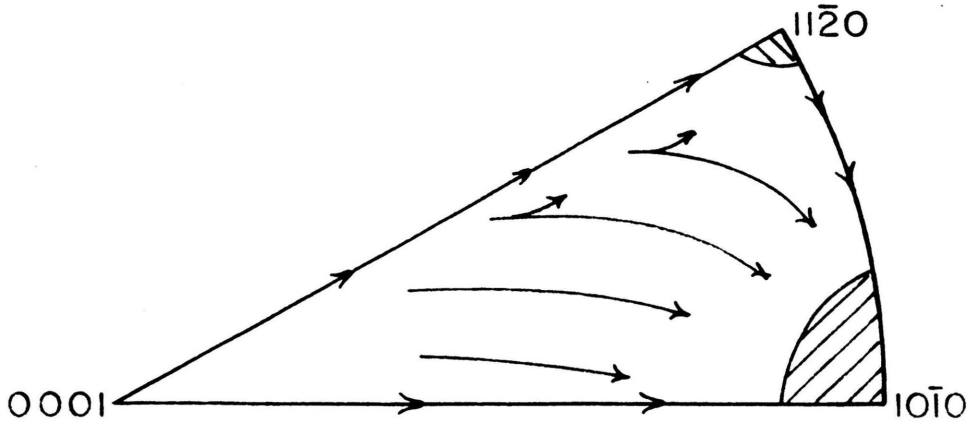


HAFNIUM — H.C.P. — C/A = 1.58

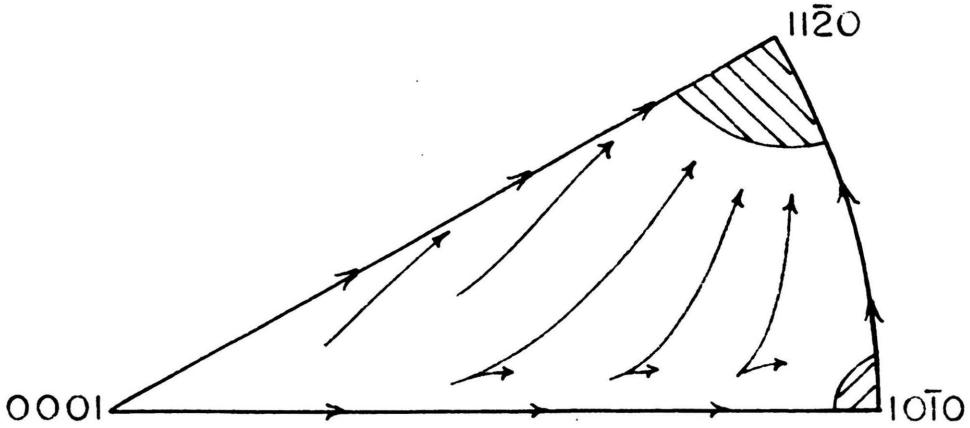
FIG. NO. 27

TENSION & COMPRESSION SLIP ROTATIONS AND TEXTURE FOR $(10\bar{1}0)$ $[11\bar{2}0]$ SLIP

TENSION



COMPRESSION

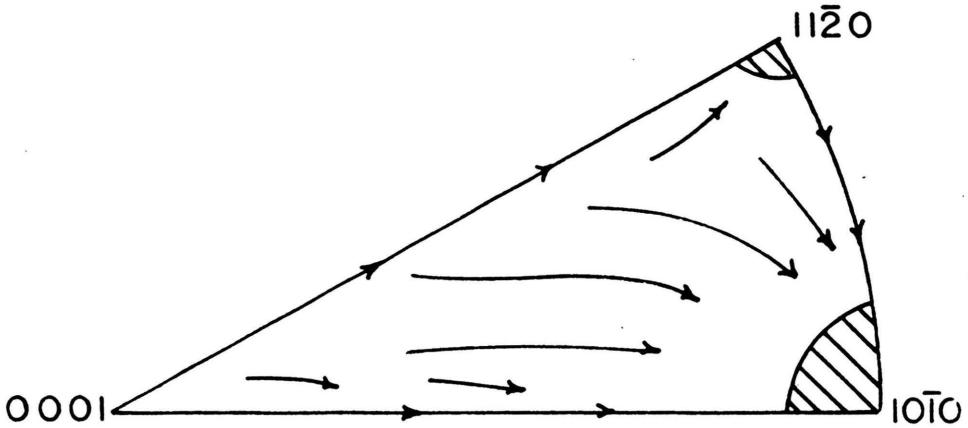


HAFNIUM — H.C.P. — C/A = 1.58

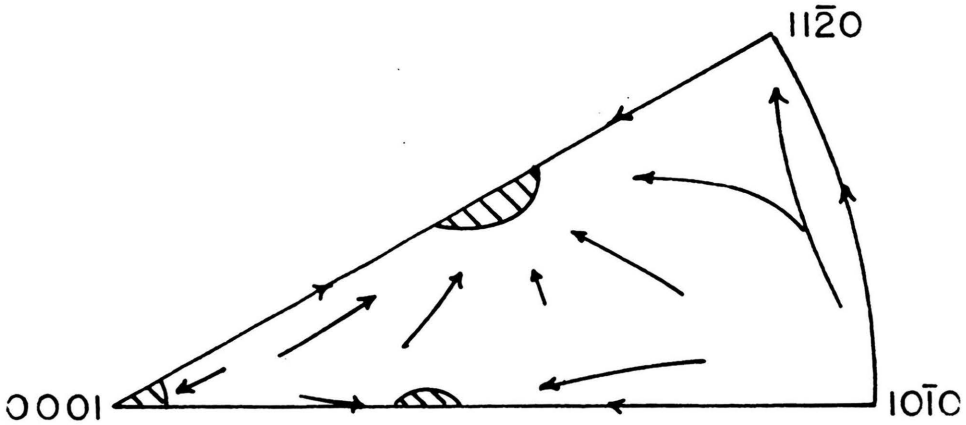
FIG. NO. 28

TENSION & COMPRESSION SLIP ROTATIONS AND TEXTURE FOR $(10\bar{1}1)$ $[11\bar{2}0]$ SLIP

TENSION



COMPRESSION

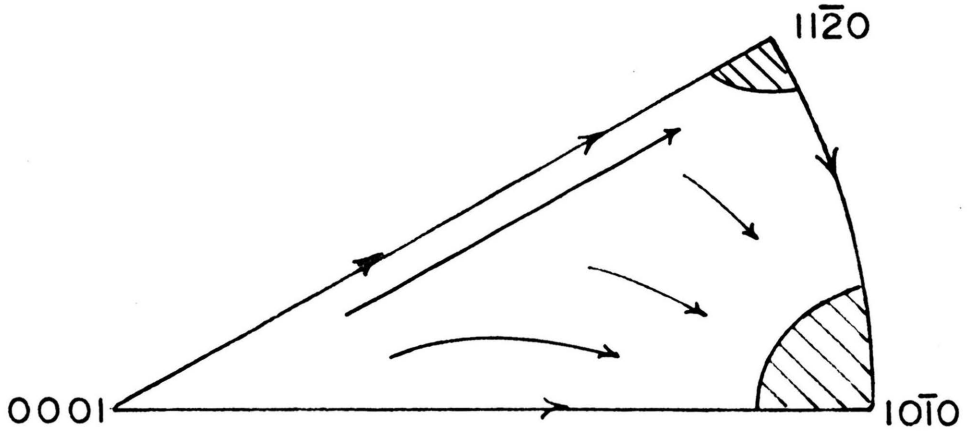


HAFNIUM — H.C.P. — C/A = 1.58

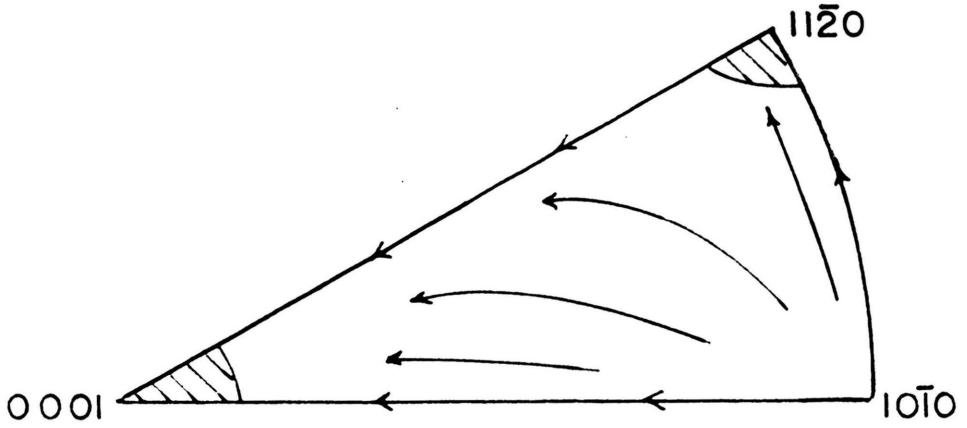
FIG. NO. 29

TENSION & COMPRESSION SLIP ROTATIONS AND TEXTURE FOR COMBINED $(0001)[11\bar{2}0]$, $(10\bar{1}0)[11\bar{2}0]$ AND $(10\bar{1}1)[11\bar{2}0]$ SLIP SYSTEMS

TENSION



COMPRESSION



HAFNIUM — H.C.P. — C/A = 1.58

$[000\bar{1}]$ $[11\bar{2}0]$ edge. This strong peak at approximately 25° away from the $[000\bar{1}]$ direction is therefore not accounted for on the basis of slip alone although the peak lies along the preferred direction of movement.

Orientation effect of twinning. The twinning observed in titanium can be grouped in two classes: $\{10\bar{1}2\}$ twinning in one class and $\{11\bar{2}2\}$ and $\{11\bar{2}1\}$ twinning in the second. Twinning reorientations in hexagonal metals are due to deformation have been indicated by Schmid and Boas[71], and Williams[72]. The most noticeable change in hafnium from other hexagonal metals of larger c/a ratios is that $\{10\bar{1}2\}$ compression twinning can no longer occur from the 0001 position. This is due to the large angle between the c axis and the $(10\bar{1}2)$ plane. $\{11\bar{2}2\}$ and $\{11\bar{2}1\}$ can and do twin from this position in titanium and it is assumed they do in hafnium. They both have similar reorientation effects, tending to move the basal plane out of the rolling plane in a $[\bar{1}\bar{1}20]$ direction. $\{10\bar{1}2\}$ twinning can occur from the $10\bar{1}0$ position but has little effect on the resulting texture.

In tension $\{1122\}$ and $\{1121\}$ twinning tend to occur from position near the 1120 point on the unit stereographic triangle. The overall effect of this is to help develop the $[\bar{1}010]$ directionality. In addition $\{1012\}$ type twinning occurs from the 0001 position also helping to develop the $[\bar{1}010]$ directionality.

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(71) E. Schmid and W. Boas, Plasticity of Crystals, (London: F. A. Hughes and Co., 1950), pp. 94-98.

(72) Williams, Loc. cit.

On the following page the slip system rotation and texture diagrams for the three operating systems are combined with the twinning reorientation effects to indicate the tension and compression texture for hafnium based on the foregoing assumptions.

Rationalization of the cold rolled texture. Cold rolling is assumed to be a tensile deformation in the rolling direction and compression deformation in the rolling plane and therefore along transverse direction (73). With such an assumption, the two textures are compatible in that when a $[\bar{1}120]$ direction occurs along the transverse axis, the compression axis, and as a result the $[\bar{1}010]$ direction will occur along the tensile axis. This $[\bar{1}010]$ directionality in the tensile axis coincides with the tensile directionality in the rolling direction which is the tensile direction.

In Figure 31 the two textures are combined to form a (0001) pole figure. In the transverse direction compressive deformations only are considered and in the tensile direction only tensile deformations are considered. At intermediate angles the relative effect is proportional to the angle away from the rolling or transverse direction. At an angle of 45° from the rolling direction in the rolling plane, compression and tension deformation are assumed, therefore, to be equal.

An area of approximately 30° from the rolling direction is forbidden for (0001) poles as a result of $\{1012\}$ twinning in tension. Such twinned poles tend to be moved toward the transverse axis.

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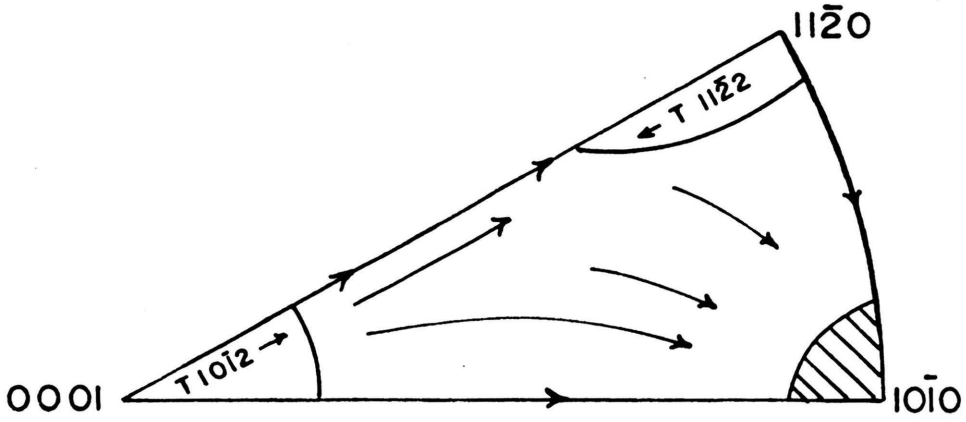
(73) F. Wever, "Textures of metals after cold deformation," Trans. AIME, Vol. 93, pp. 51-75, 1931.

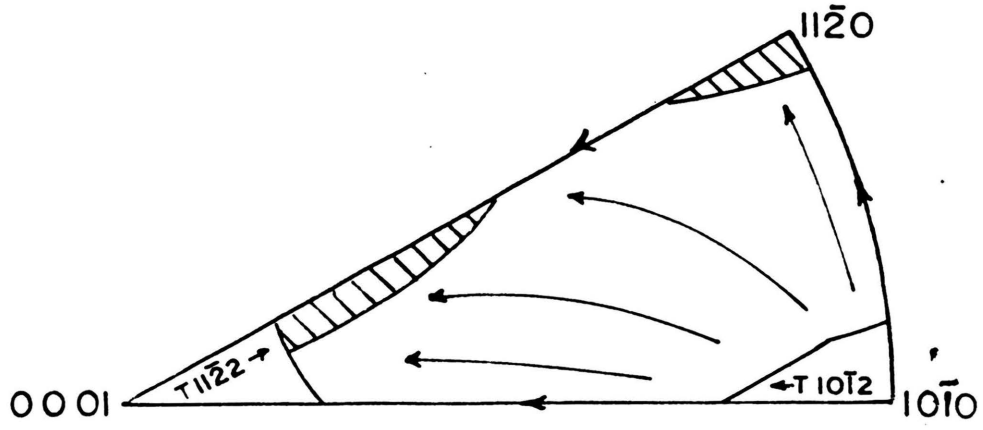
FIG. NO. 30

TENSION & COMPRESSION ROTATIONS AND TEXTURE FOR COMBINED SLIP AND TWINNING

TENSION



COMPRESSION

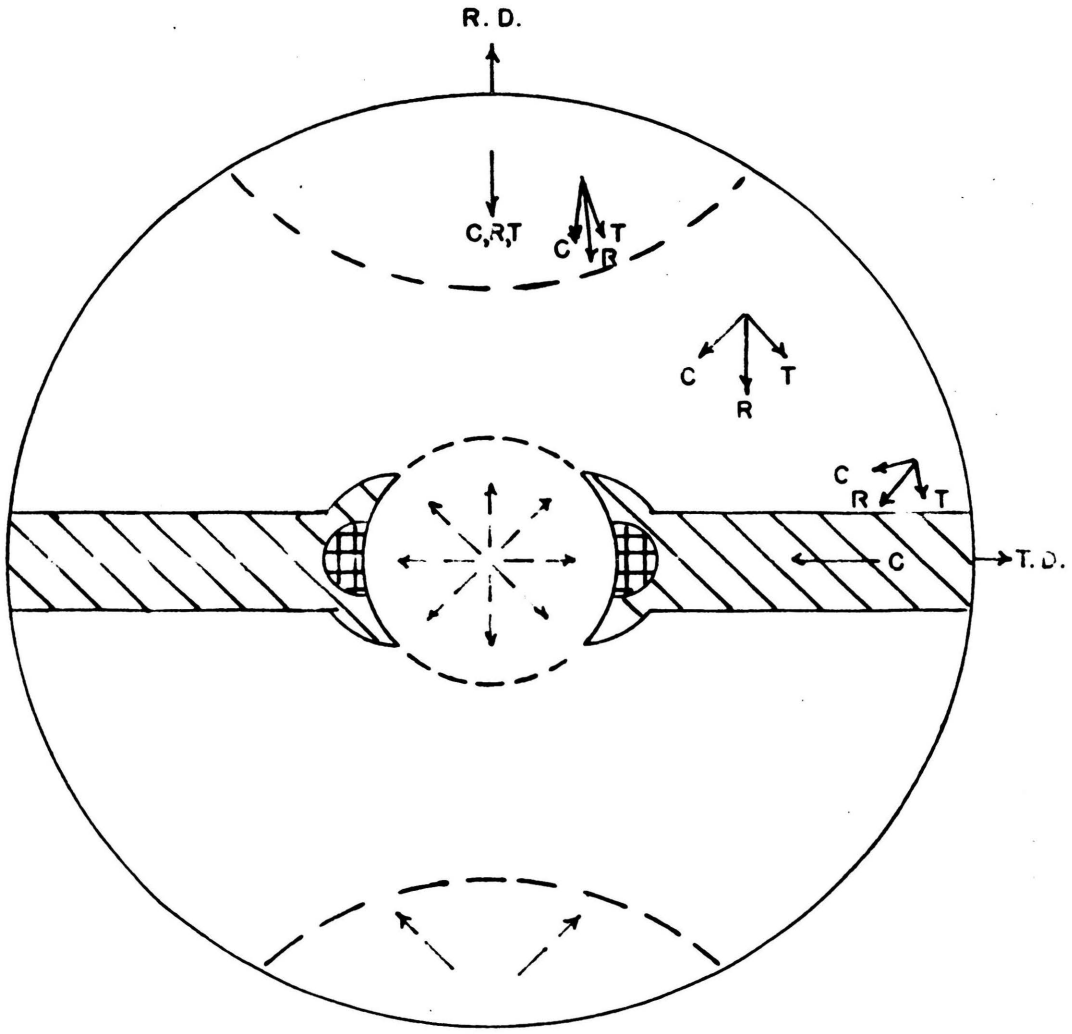


HAFNIUM — H.C.P. — C / A = 1.58

FIG. NO. 31

THEORETICAL (0001) POLE FIGURE

COMBINED TENSION AND COMPRESSION DIAGRAM



DASHED CIRCLES INDICATE FORBIDDEN AREAS

HAFNIUM — H.C.P. — C /A = 1.58

Compression twinning also prohibits (0001) poles from being in the center of the pole figure. The actual area is uncertain theoretically because it is not known whether $\{11\bar{2}1\}$ or $\{11\bar{2}2\}$ twinning will occur and because no relationship between the critical shear stress for (0001) slip and $\{11\bar{2}1\}$ type twinning has been assumed. From the experimental data it can be assumed that the forbidden area extends approximately 25° from the rolling plane normal.

Discussion. Using the Calnan and Clews method; it is possible, on the basis of the experimentally determined texture, to adjust the boundaries between the operating slip systems and arrive at a comparative ratio of the critical shear stress values for the various systems. This adjustment of system boundaries also makes it possible to fit the theoretical texture specifically to the experimentally determined texture. This procedure of boundary adjustment was proposed by Williams[74] and applied to the texture analysis of titanium[75].

Calnan and Clews, in their original work[76] assumed that the critical shear stress for all systems was of the same order of magnitude and made no boundary adjustment. The present author feels that this is the preferred procedure for several reasons. In that ratio between critical shear stresses is not necessarily a constant value, any value for a given sample is apt to be inaccurate when applied to a

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(74) Williams, Loc. cit.

(75) Williams and Eppelsheimer, Loc. cit., (Ref. 3).

(76) Calnan and Clews, Loc. cit., (Ref. 36).

different sample. As an example, Williams and Eppelsheimer indicate a ratio of $C_s0001 = 1.02\ C_s10\bar{1}0$. Anderson et all[77] have experimentally determined that the ratio should be closer to $C_s0001 = 2.5\ C_s1010$. A second reason that the author prefers not to adjust the diagram to the experimental data is that an adjusted diagram becomes representative of only the one experimental sample. As long as the correct assumptions are originally made with respect to which systems can operate, a general diagram can include any experimentally determined texture without regard to the amount of deformation or rolling procedure. Workers subsequent to Williams and Eppelsheimer[78] have found large variations in the cold rolled texture of titanium[79].

Except for the point mentioned above, it is observed that the preceeding texture analysis for hafnium is similar to that for titanium and as might be expected the resulting texture is much the same. The resultant texture diagram for hafnium can be described as a (0001) $[10\bar{1}0]$ texture rotated at various angles from the rolling plane. As can be noted from the diagram the tendency is for the poles to be crowded toward the twinning limit but poles all along the transverse axis are entirely possible.

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(77) Anderson, Jillson and Dunbar, Loc. cit.

(78) Williams and Eppelsheimer, Loc. cit.

(79) J. H. Keeler and A. H. Geisler, "Preferred orientations in rolled and annealed titanium," J. of Metals, AIME, Vol. 7, pp. 80-90, (Feb.), 1956.

CHAPTER VI

THE RECRYSTALLIZATION TEXTURE OF HAFNIUM

The recrystallization textures of Ti and Zr have been reported by early workers to be essentially a (0001) $[\bar{1}120]$ tilted at various angles in the transverse direction [80, 81, 82]. The recrystallization texture of hafnium might be expected to be similar in that the cold rolled texture of hafnium is so similar to titanium and zirconium.

Recent investigators have modified the basic (0001) $[\bar{1}120]$ texture noted above. Under certain recrystallization conditions, the directionality of the texture appears to rotate [83, 84]. This rotation is evidenced by an appreciable angle between the tilt axis and the rolling direction which are coincident in the basic texture. Such a texture is characterized by a splitting of the peaks found in the basic texture. This angle of rotation between the axis of tilt and the rolling direction appears to be variable and a possible function of

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(80) D. N. Williams and D. S. Eppelsheimer, "Recrystallization texture of titanium," Zeit. fur Metallkunde, Vol. 44, p. 360, 1953.

(81) C. J. McHargue and J. P. Hammond, "Preferred orientations in iodide titanium," J. of Metals, AIME, Vol. 5, p. 57, (Jan.), 1953.

(82) R. K. McGeary and B. Lustman, "Preferred orienations in zirconium," J. of Metals, AIME, Vol. 3, p. 994, (Nov.), 1951.

(83) Keeler, Hibbard and Decker, Loc. cit.

(84) Keeler and Geisler, Loc. cit.

both temperature and time at temperature where the temperature is not constant.

A. EXPERIMENTAL PROCEDURE

This investigation was based on cold rolled strips 0.004" thick resulting from 95% reduction by cold rolling of the arc melted material. The principle impurity was approximately 3% zirconium. The recrystallization range for similar material was reported as being between 700°C and 800°C[85] although there was some indication that it might be a little higher[86].

The recrystallization annealing was done in a vacuum retort furnace. The retort consisted of a glazed porcelain tube. The sealed end of the tube extended into an electric resistance furnace. The unsealed end contained a thermocouple well and the vacuum connection. The vacuum connection could be removed quickly and the samples were loaded and unloaded through this connection.

The annealing procedure consisted of bringing the furnace up to the desired temperature as recorded by the thermocouple in the retort. The furnace was allowed to come to equilibrium with the vacuum pump operating with the current supplied to the furnace being adjusted until there was no appreciable variation in furnace temperature. The pump was then shut off; the connection opened; the sample inserted as close

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(85) Litton, Loc. cit.

(86) Roth, Loc. cit.

to the tip of the thermocouple tube as possible. The retort was again sealed and the pump started. It was observed that there was less than 5°C temperature drop within the furnace during the loading process and since the samples were 1/2" square and only 0.005" thick, it was assumed that they came to temperature rapidly. At the end of the one hour recrystallization annealing time the connection was broken and the samples removed. Since the samples cooled very quickly upon removal, there was only a very thin oxide film. The vacuum in the retort furnace was of the order of 10 microns. The temperature variation was approximately $\pm$ 5°C.

Samples were annealed at 500°, 650° and 800°C; each for one hour.

Although the oxide film on the samples after annealing was very thin it was found to be very hard and tenacious. The film appeared to resist chemical attack by ordinary chemical reagents. To remove the film, mechanical polishing was used after which approximately 0.001" was etched from the surface of the sample using a solution of 10% conc. HF, 10% conc. HCl, 10% HNO_3 conc. and 70% H_2O. Hardness and preferred orientation determinations were made on the resulting bright etched surfaces.

Hardness determinations using exactly the same procedure as outlined for the cold rolled material were made on the recrystallization samples.

The 800° annealed sample was electrolytically etched in a solution of one part 70% perchloric and 6 parts glacial acetic acids

according to a procedure outlined for zirconium[87].

The preferred orientation of all three samples was determined using the procedure outlined in the appendix. Reflection pole figures of (0002) and $(10\bar{1}1)$ planes were prepared. The outer portion of the $(10\bar{1}0)$ pole figure was also determined using the procedure described for cold rolled transmission samples.

B. EXPERIMENTAL RESULTS

Figure 32 shows graphically the hardness of the three annealed samples as compared to the hardness of the cold rolled material. On the basis of hardness the 650° annealed sample would appear to be substantially recrystallized. As indicated on page 43, the hardness values shown have a possible precision variation of ± 20 D.P.H.

The microstructure of the sample annealed one hour at 800°C is shown in Figure 33. It is immediately evident that the grain size of the recrystallized material is much smaller than the original material which had grains of the order to several millimeters in diameter.

Using the formula[88]:

$$D = \frac{1.075}{M}\sqrt{\frac{A}{n}}$$

where D equals the distance between hexagonal grain flats, M equals magnification, A equals field area, and n equals number of grains in

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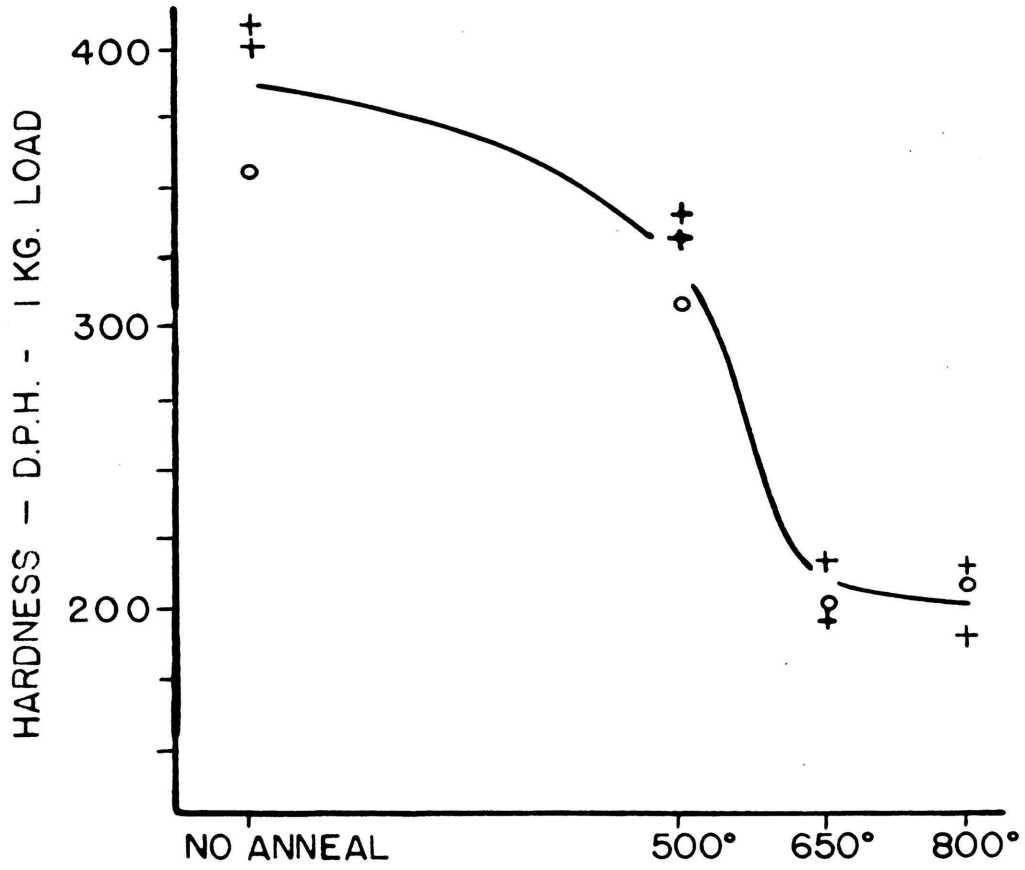
(87) H. P. Roth, "Metallography of zirconium," Metal Progress, ASM, Vol. 58, p. 709, (Nov.), 1950.

(88) F. J. Dunkerley, F. Pledger, V. Damino and J. Fulton, "Grain growth and recrystallization characteristics of zirconium," Trans. AIME, Vol. 191, p. 103, 1951.

FIG. NO. 32

HARDNESS VS ANNEALING TEMPERATURE

HAFNIUM STRIP – 95% COLD REDUCED & ANNEALED



\+ = M.S.M. HARDNESS TESTER

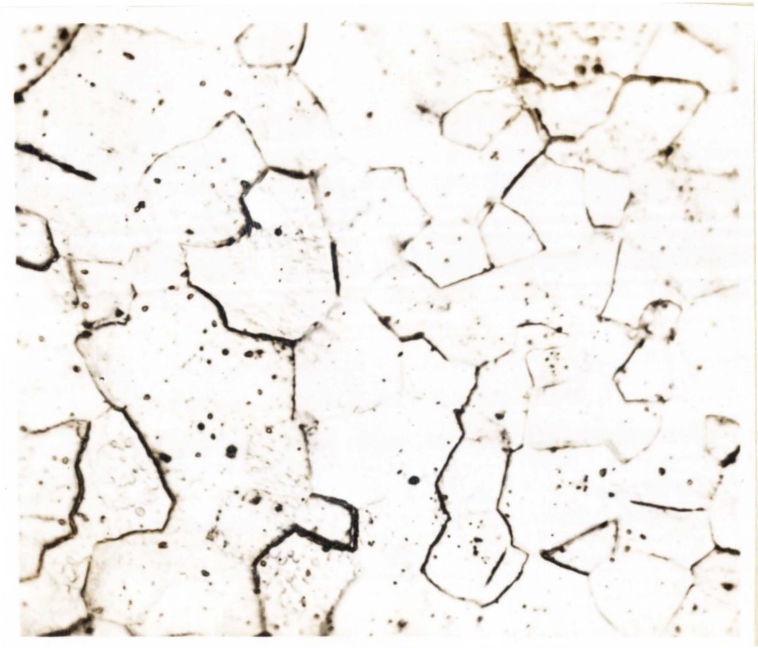
o = U.S.B.M. HARDNESS TESTER

NOTE: ALL SAMPLES ANNEALED ONE HOUR

FIG. NO. 33

PHOTOMICROGRAPH OF

RECRYSTALLIZED COLD ROLLED HAFNIUM STRIP



MAGNIFICATION : 250 X

ETCHING: ELECTROLYTIC ETCH AT 16 VOLTS, 0.2 AMPS / CM^2 IN AN ELECTROLYTE OF ONE PART 70% PERCHLORIC ACID TO SIX PARTS GLACIAL ACETIC ACID

the given area; the average grain size is found to be equal to 0.0425 mm. It is interesting to note that with this relatively small grain size several thousand grains are exposed to the x-ray beam as a result of the sample oscillation in pole figure determination.

The $(10\bar{1}1)$ pole figures were made for the three samples and are shown on the following pages. The 500° and 650° annealed samples do not appear to have an ideally resolved texture; the 500° sample appears to have a texture closer to the cold rolled orientation than the 650° annealed sample. The $[11\bar{2}0]$ directionality is evident only in the 800° sample. On the basis of the $(10\bar{1}1)$ pole figure for the 800° sample the recrystallized texture would appear to be a (0001) $[11\bar{2}0]$ texture rotated approximately 5° in the transverse direction. The (0002) pole figure did not resolve the expected tilt of the basal poles in the transverse direction. It merely indicated a large concentration of basal poles in the center of the pole figure diagram. The split structure sometimes noted in zirconium and titanium was not evident in the $(10\bar{1}1)$ pole figure. The peaks in the transverse direction were rather large indicating that the split texture may merely have been unresolved. In an effort to determine if the 800° anneal sample had a true $[11\bar{2}0]$ directionality, an effort was made to run the outer area of the (1010) transmission pole figure. The major difficulties associated with transmission pole figure determination of the cold rolled samples were also encountered with recrystallized samples. As can be noted in Figure 37, the contrast is not good, but this $(10\bar{1}0)$ pole figure would tend to indicate that the recrystallization texture has a $[11\bar{2}0]$ direction.

FIG. NO. 34

INNER 60° PORTION OF

(10$\bar{1}$1) POLE FIGURE

HAFNIUM STRIP 95% COLD REDUCED

ANNEALED ONE HOUR AT 500° C.

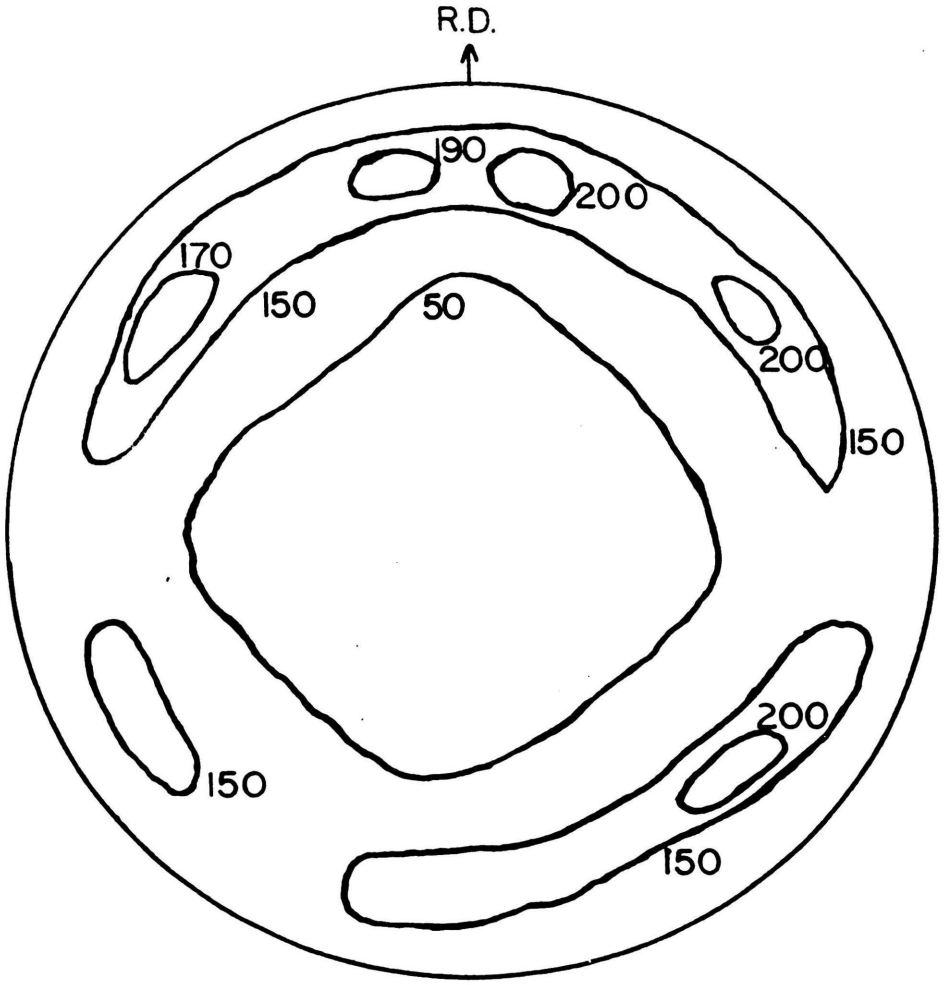


FIG. NO. 35

INNER 60° PORTION OF

(10$\bar{1}$1) POLE FIGURE

HAFNIUM STRIP 95 % COLD REDUCED

ANNEALED ONE HOUR AT 650° C.

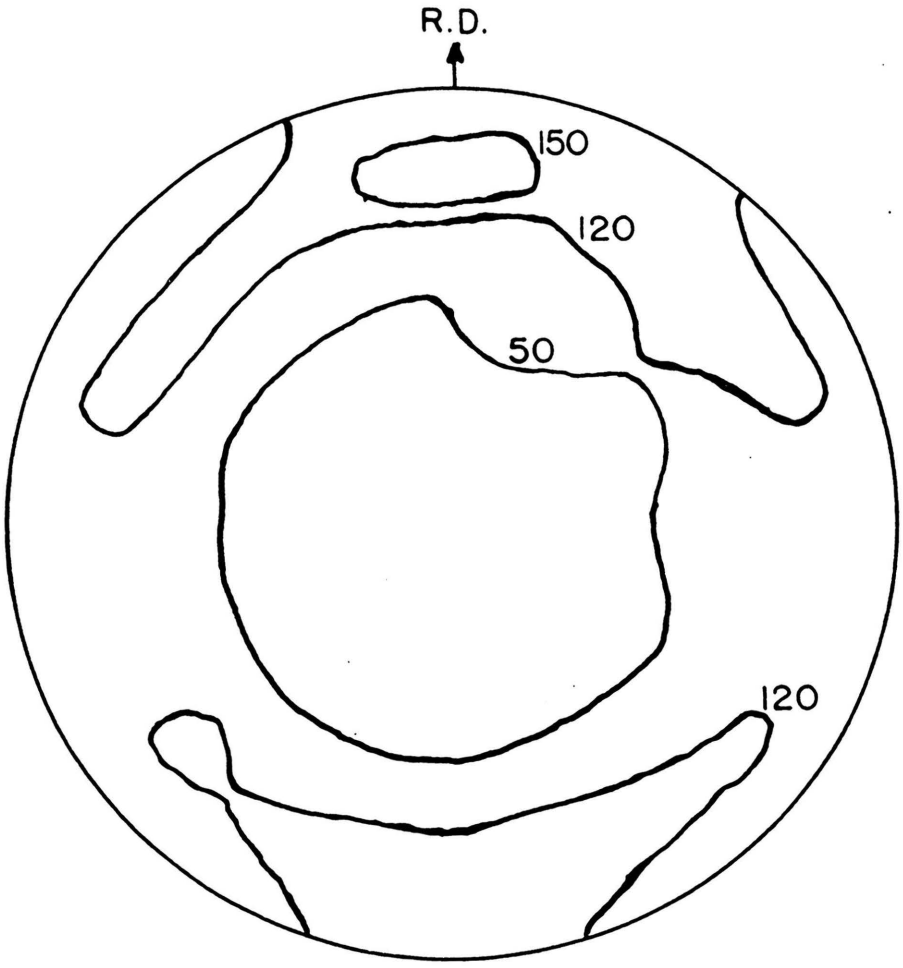


FIG. NO. 36

INNER 60° PORTION OF

(10$\bar{1}$1) POLE FIGURE

HAFNIUM STRIP 95% COLD REDUCED

ANNEALED ONE HOUR 800° C.

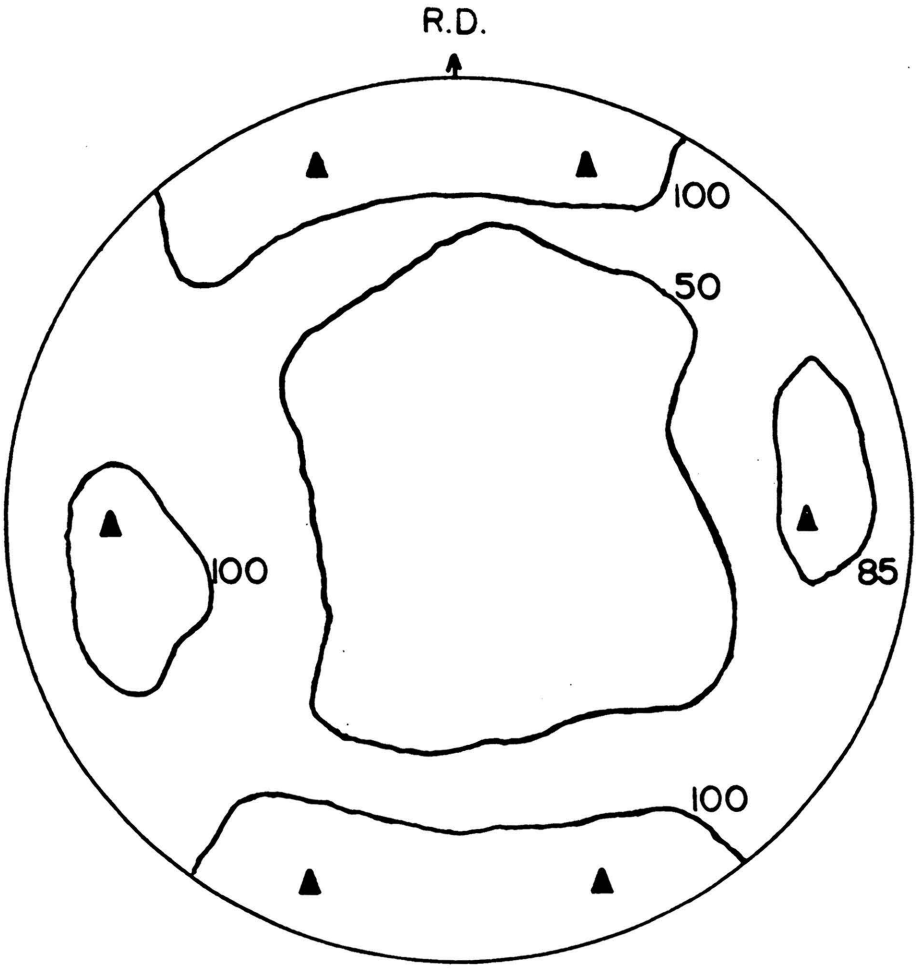
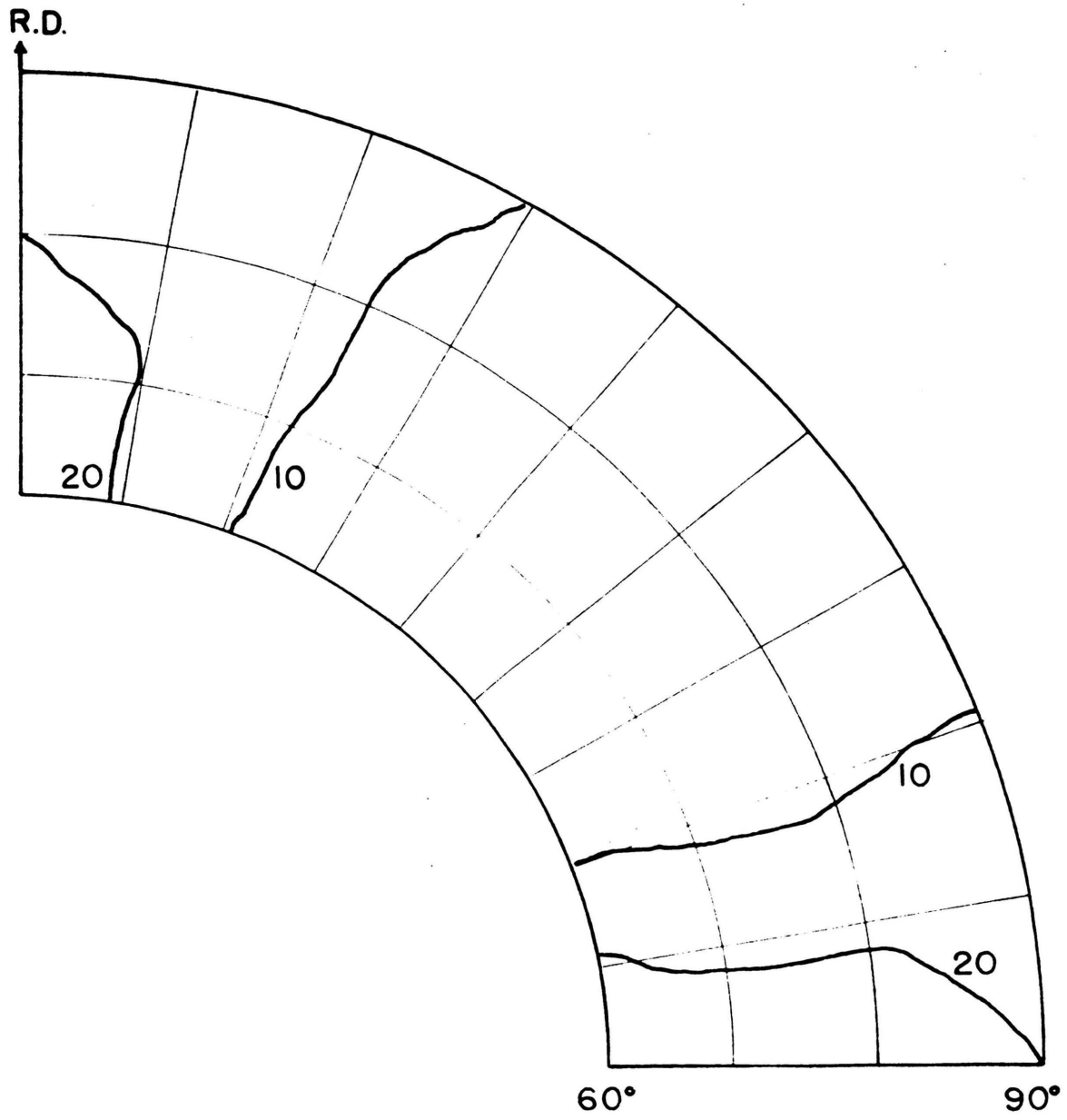


FIG. NO. 37

(10$\bar{1}$0) POLE FIGURE

TRANSMISSION SEGMENT

HAFNIUM STRIP 95% COLD REDUCED
ANNEALED ONE HOUR AT 800°C IN VAC.



Assuming the same precision accuracy as for the cold rolled texture, the recrystallization texture for hafnium strip containing 3% Zr cold rolled 95% reduction and annealed for one hour at 800°C can be described as a (0001) $[1\bar{1}20]$ texture rotated 5° ± 2° in the transverse direction.

CHAPTER VII

ACTIVATION ENERGY FOR RECRYSTALLIZATION OF HAFNIUM STRIP

Several investigators have studied the activation energy of materials which have a pronounced change in texture upon recrystallization by x-ray diffraction spectrometer techniques. The technique devised by Decker and Harker[89] for copper was also successfully applied to silver by Tangri and Eppelsheimer[90]. Essentially, this technique involves setting up the diffraction spectrometer to record the intensity of a Bragg reflection associated with the recrystallization texture. The comparative intensity of this reflected peak is then a measure of the degree of recrystallization of the sample. The results reported for this technique appear to have an accuracy greater than techniques utilizing changes in hardness, tensile strength etc.[91, 92].

The investigation of the textures of hafnium indicated a

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(89) B. F. Decker and D. Harker, "Activation energy for recrystallization in rolled copper," J. of Metals, AIME, Vol. 188, p. 887, (Jun.), 1950.

(90) K. K. Tangri and D. S. Eppelsheimer, "Activation energy for recrystallization in rolled silver," Missouri School of Mines Technical Bulletin No. 83, Jan., 1954.

(91) M. Cook and T. L. L. Richards, "Observations of the rate and mechanism of recrystallization in copper," Jour. of Inst. of Metals, Vol. 73, p. 1, 1947.

(92) N. Thorley, "Calculation of the activation energies of recovery and recrystallization from hardness measurements on copper," Jour. of Inst. of Metals, Vol. 77, p. 141, 1950.

change from a [10$\bar{1}$0] cold rolled directionality to a [11$\bar{2}$0] directionality in the recrystallized material. This change in texture affords the means for making degree of recrystallization measurements.

The Norelco pole figure fixture described in the appendix provided a unique method of determining degree of recrystallization which would appear to be more refined than the previously described techniques. As can be noted in Figure 38, rotation through 360° at a ϕ angle of 45° will cover all the (10$\bar{1}$1) poles rotated at an angle of 45° from the rolling plane normal. This rotation will record all the (10$\bar{1}$1) peaks in either the cold rolled or recrystallized texture. It is immediately evident that measurement of the peaks in the rolling direction serves no purpose, but that the peaks in the transverse direction are characteristic of the recrystallized structure and peaks approximately 40° from the transverse direction are characterisitic of the cold rolled texture. Therefore the basis of the technique used was to compare the relative heights of these two sets of peaks.

It would appear that scanning a number of peaks plus continuous oscillation of the sample would constitute a significant improvement over the previously employed technique.

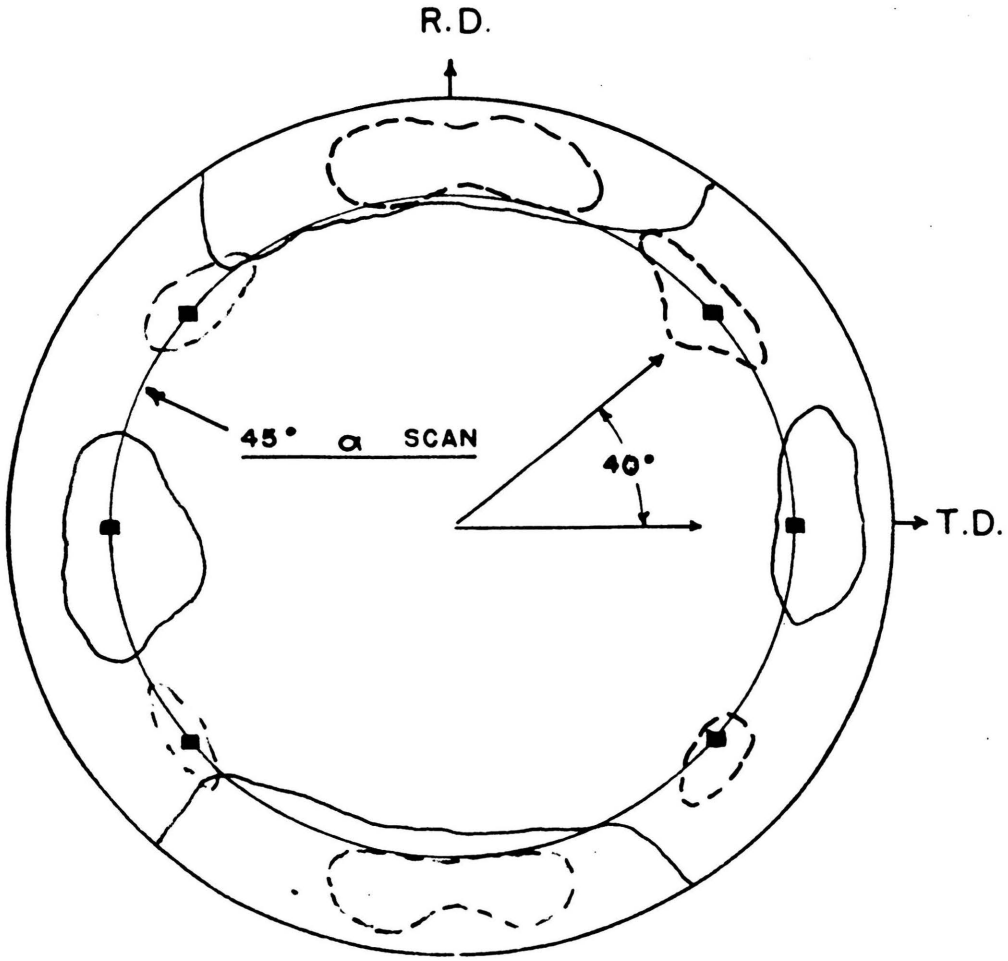
A series of samples was sectioned from a cold rolled strip and heated in a lead bath for various lengths of time at 600°, 650°, 700°, 750° and 800°C. The lead bath provided freedom from excessive oxidation and since the samples were only 0.006" thick they cooled very rapidly upon being withdrawn from the bath. Because of this, the times at the recrystallizing temperatures were controlled very closely. The

FIG. NO. 38

POSSIBLE PEAKS IN 45° α SCAN

DASHED LINES INDICATE COLD ROLLED TEXTURE, SOLID LINES RECRYSTALLIZATION TEXTURE OF HAFNIUM STRIP COLD REDUCED 95% AND HEATED ONE HOUR AT 800°C.

INTENSITY READINGS MADE ■ POINTS



COMPOSITE C.R. & ANNEALED
(10$\bar{1}$1) POLE FIGURES

temperature of the bath was measured by a thermocouple immersed in the bath and the temperature was found to vary cyclically by approximately ± 5°C. The chromel-alumel thermocouple used had previously been calibrated using a Leeds and Northrup precision potentiometer. Subsequent to the preliminary calculations a series of samples was also run at 775°C. After being removed from the lead bath the surface was mechanically polished and then approximately 0.001" was etched from the surface.

The samples were placed on the stage of the pole figure fixture, the 2θ angle set for $(10\bar{1}1)$ reflection, and the stage then rotated through the complete 360° α rotation at a rate of 20° of α rotation per minute. The samples were also oscillated in their own plane in order to obtain an intensity readings as representative of the entire sample as possible. It is realized that the rotation may not go through the center of the appropriate $(10\bar{1}1)$ peaks and as a result the highest intensity value may not be recorded. Since such a deviation would be constant for a given set of samples it will have no effect as far as the ultimate results are concerned. A typical set of recorded intensity charts for three samples heated at various times at 775°C is shown in Figure 39 on the following page. The check points indicated by the squares in Figure 38 are measured above the appropriate points noted along the horizontal axis in Figure 39.

The complete set of average peak heights measured for all the samples run is recorded in Table 3. The ratio of the intensity of the cold rolled peak to the recrystallized peak height is shown in the

FIG. NO. 39

α SCAN OF 45° POLES

PARTIALLY RECRYSTALLIZED HAFNIUM STRIP

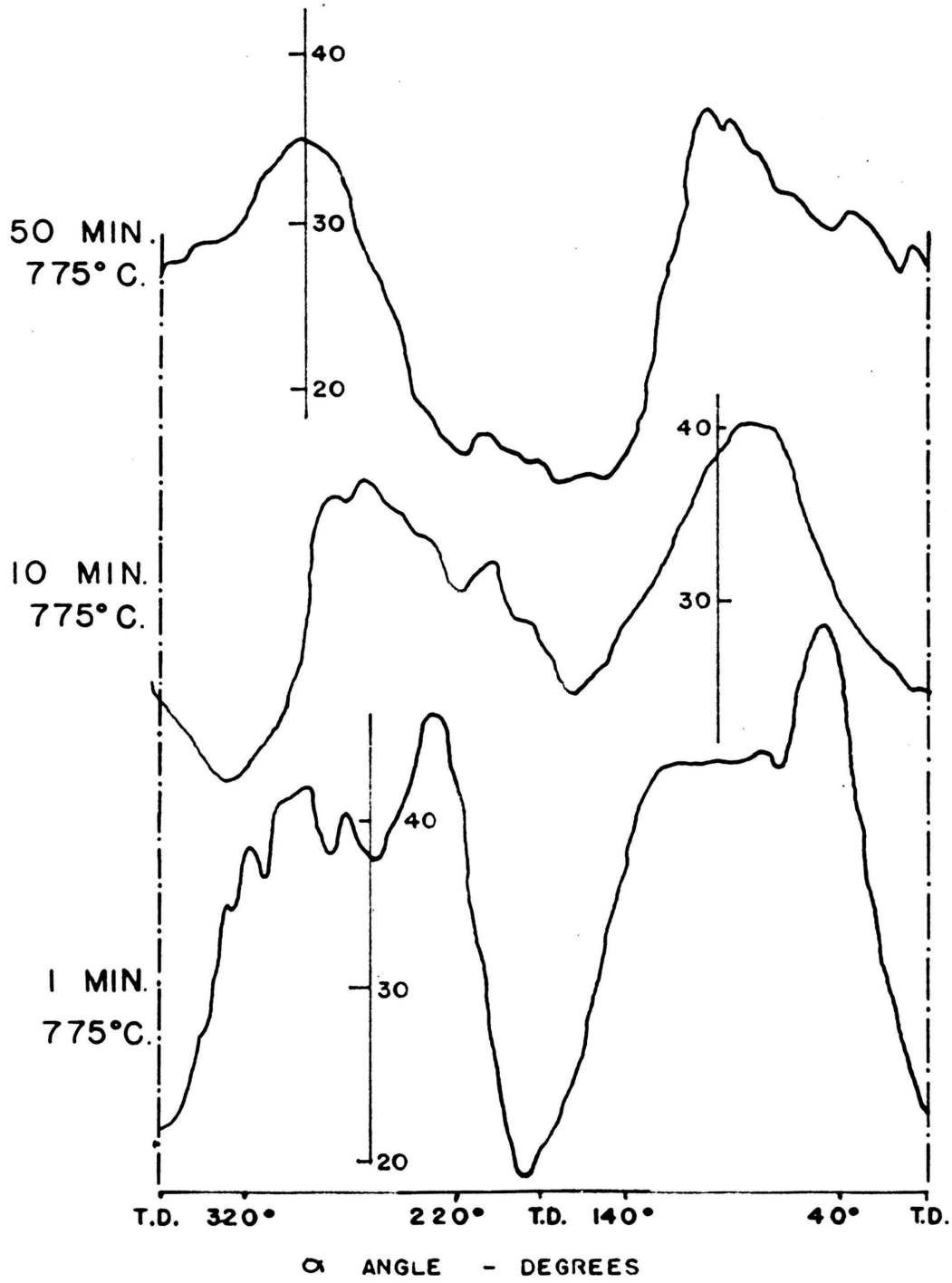


TABLE III

PEAK HEIGHT RATIOS FOR RECRYSTALLIZED SAMPLES

Sample	Cold-Rolled Peak	Recrystallized Peak	Ratio
As Cold Rolled	48	23	2.10
600°-10 min.	40	20	2.00
600°-100 min.	42	23	1.83
650°-10 min.	46	25	1.84
650°-100 min.	50	29	1.72
700°-10 min.	48	26	1.84
700°-100 min.	31	23	1.34
750°-1 min.	51	28	1.82
750°-10 min.	45	29.5	1.52
750°-100 min.	33	36	.92
775°-1 min.	42	21	2.00
775°-10 min.	30	24	1.25
775°-50 min.	24	21	1.14
775°-100 min.	20	22	.91
800°-1 min.	34	19	1.79
800°-10 min.	27	28	.96
800°-20 min.	18	20	.90
800°-50 min.	22.5	25	.90

right hand column. This ratio is plotted against time in Figure 40. It is evident, upon examination of the strip charts, that an assumption of a precision measurement of more than ± 0.05 in ratio intensities would be presumptous. It is noted that the ratio of cold rolled texture intensities is approximately 2.0 and the intensity ratios seem to become constant at about 0.9. It can be assumed that a ratio of 0.9 indicates complete recrystallization. On the basis of these two ratios, 1.4 was chosen as the ratio of approximately 50% recrystallization. Whether it is exactly 50% is, as will be pointed out later, of no significance. However it is relatively safe to assume that samples with an intensity ratio of 1.4 contain the same degree of recrystallization.

The rate of recrystallization is given by the equation:

$$\text{Rate} = Ae^{\frac{-Q}{RT}}$$

where Q is an activation energy, R the gas constant, T the absolute temperature and A a constant. From the above:

$$\frac{1}{t} = A'e^{\frac{-Q}{RT}}$$

where t is the time necessary for a given amount of recrystallization to take place at temperature T. Then:

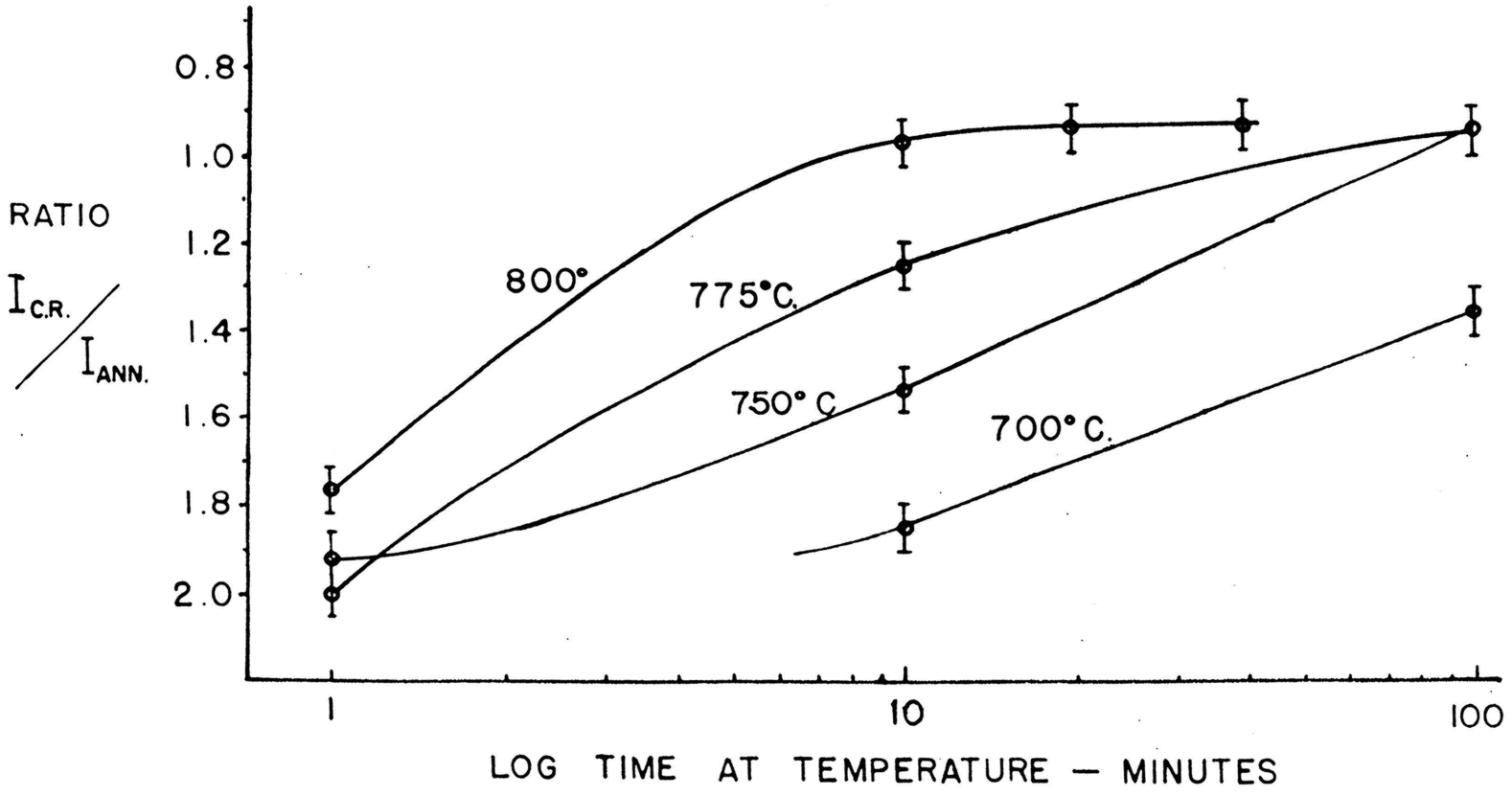
$$\frac{d\ \frac{1}{T}}{d\ (\log t)} = \frac{R(2.303)}{Q}$$

The expression on the left hand side of the above equation represents the slope of a graph of reciprocal of absolute temperature versus the time necessary for a given amount of recrystallization. From the graph

FIG. NO. 40

PEAK HEIGHT RATIO VS TIME AT TEMPERATURE

ISOTHERMAL RECRYSTALLIZATION CURVES FOR HAFNIUM STRIP 95% COLD REDUCED



in Figure 40 it is evident that a point near 50% recrystallization would give the most accurate value for t for the above equations as their slopes are essentially linear near these midpoints. Small amounts or nearly completely recrystallization would give poor t values. The times (t) corresponding with an intensity ratio of 1.4 are plotted in Figure 41 against the reciprocal of the temperature of the corresponding recrystallization temperature.

The slope of the curve in Figure 41 is found to be 0.0000607. This value gives an activation energy for the hafnium strip containing 3% Zr of 75,400 cal/gm mole of 75.4 K/gm mol.

If any possible variation in the intensity ratio was consistent; all positive or all negative, it would have very little effect on the absolute value of activation energy. If however the variation was inconsistent and amounted to $\pm$ 0.1, the resulting variation in the activation energy could be as much as 20%. In that it would be difficult to know whether the variation is consistent or not, it might be best to assume that it is unpredictable and not consistent. This would indicate that the above figure may vary by as much as $\pm$ 15 K/gm mol.

The calculated activation energy therefore becomes 75 $\pm$ 15 K/gm mol.

This investigation also tends to verify the recrystallization range of 700-800°C found by Litton for hafnium containing 3% Zr (93). Roth reports a recrystallization temperature of 1100°C for 65% cold

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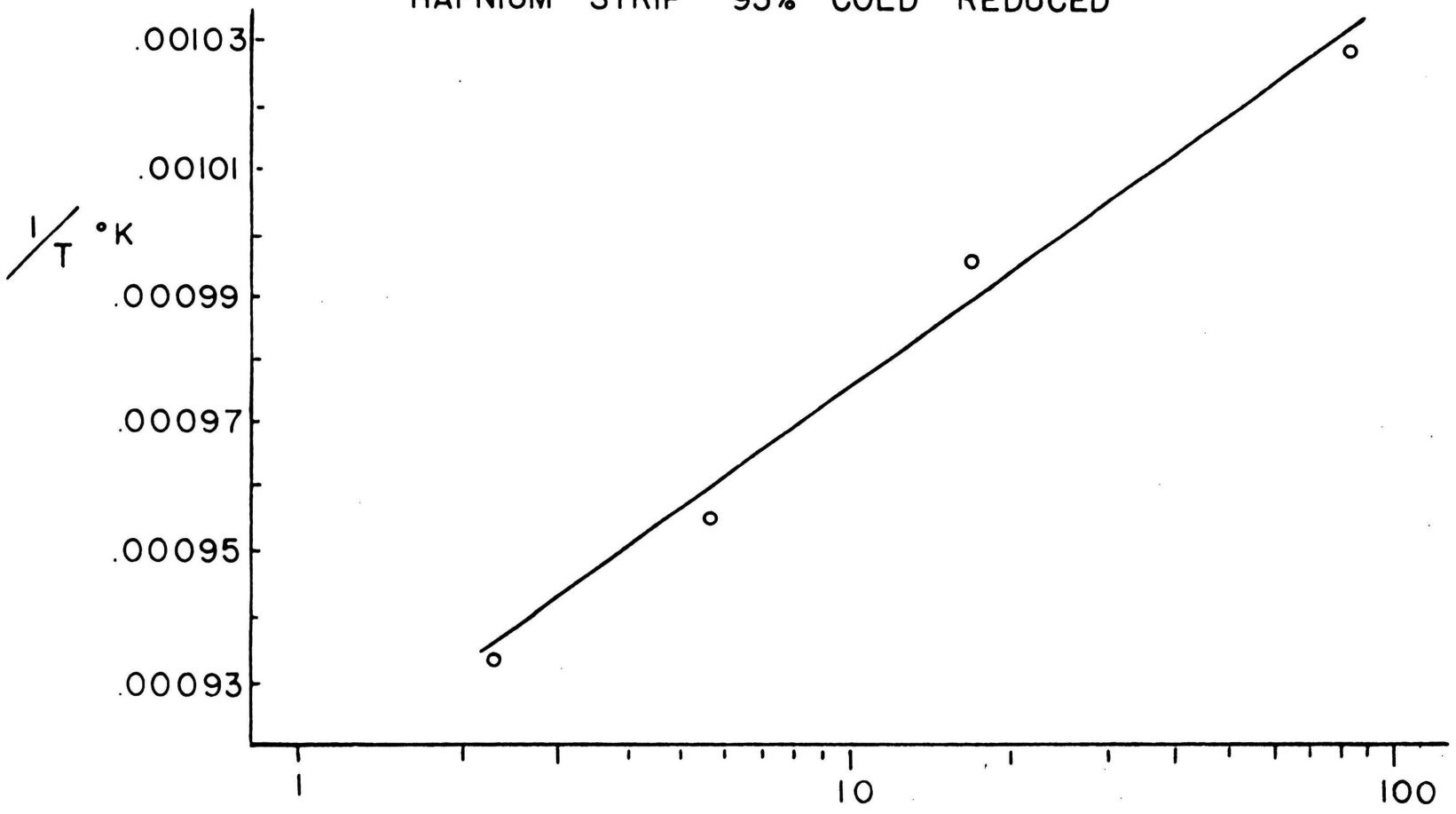
(93) Litton, Loc. cit.

FIG. NO. 41

RECIPROCAL OF ABSOLUTE TEMPERATURE VS. TIME FOR 50% RECRYSTALLIZATION

HAFNIUM STRIP 95% COLD REDUCED



reduced material (94); approximately the same cold reduction as used by Litton. Although Roth's material contained only 1% Zr, it is difficult to rationalize the vast difference in recrystallization temperatures on the basis of this relative small difference in chemical composition. This, however, is the only apparent explanation.

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(94) Roth, Loc. cit.

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A P P E N D I X

APPENDIX

X-RAY TECHNIQUE FOR DETERMINATION OF PREFERRED ORIENTATION

A. STEREOGRAPHIC PROJECTION

The orientation of crystallographic planes in a metallic sheet material is essentially a three dimensional phenomenon. A stereographic projection provides a simple way in two dimensions to record such orientation data. Barrett has provided a clear explanation of the role of stereographic projection in orientation determination (95).

Figure No. 42, taken from Barrett indicates the relationship between the sample, the Debye ring and the projection plane. The reflecting plane noted in the diagram will cause a diffraction spot at point S on the Debye ring. It can be observed that if the reflecting plane were rotated around an axis coincident with the x-ray beam, the diffraction spot would move about the Debye ring. The basic element of a stereographic projection is line normal to the reflection plane. The reflection plane is considered to be at the center of a sphere and such a normal to the reflecting plane will intersect the sphere as noted by point P in the diagram. As can be further observed in the diagram, point P is projected from a point where the sphere, the x-ray beam and the Debye plane intersect at a point P_1 on the projection plane which abuts the sphere on the opposite side and also normal to the x-ray beam. These normals from the reflection planes to the re-

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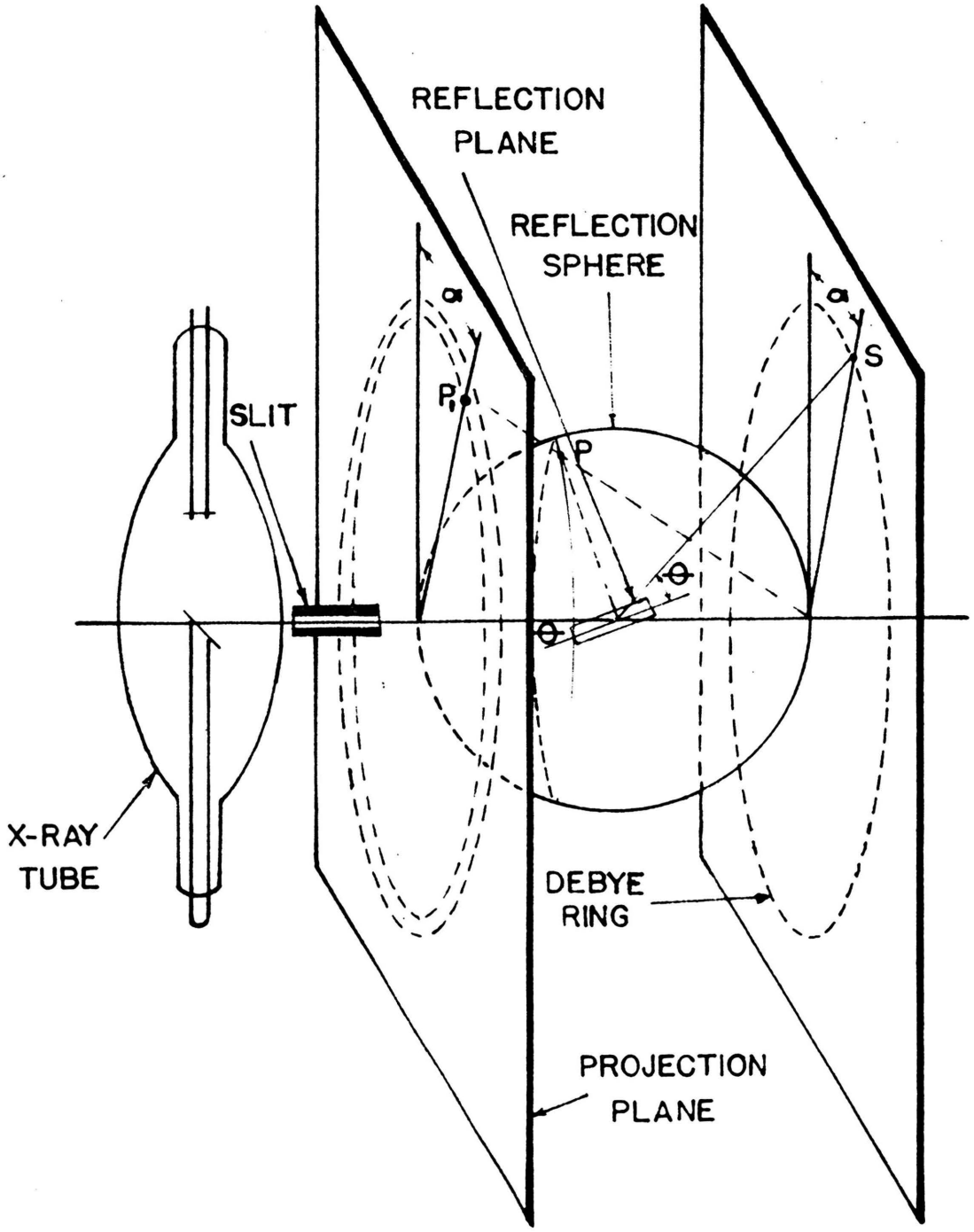
(95) C. S. Barrett, Loc. cit.

FIG. NO. 42

RELATIONSHIP BETWEEN REFLECTING PLANE DIFFRACTED BEAM AND STEREOGRAPHIC PROJECTION



flection sphere are called "Poles" in texture work, hence the name "Pole Figure" for the projection of their intersection with the sphere to the projection plane.

As the spot S moves around the Debye ring, point P_1 moves around a corresponding circle on the projection plane. It can be observed that the pole of a reflecting plane normal to the x-ray beam will be projected to the center of the projection plane and the pole of a plane that lies parallel to the x-ray beam would be at the edge of the projection plane. Therefore it follows that the angle which the reflecting plane makes with the x-ray beam is expressed by an angular distance, ϕ, away from the center of the projection plane. The α angle noted on the diagram is the reference relative to the rotation of the plane around the axis of the x-ray beam. With these two references any point of the pole figure represents a geometric relationship between the reflecting plane and the x-ray beam.

In polycrystalline material it is normally considered that a great number of reflecting planes with a given 2θ diffraction angle intersect the center of the reflection sphere. In a randomly oriented powder sample this would mean that the reflecting plane poles would intersect the reflection sphere more or less uniformly over its entire surface. As a result, the projection plane would be uniformly populated with points each indicating an individual plane orientation with respect to the incident x-ray beam.

In texture work using the Schulz reflection technique[96], the

(96) L. G. Schulz, "A direct method of determining preferred orientation of a flat reflection sample using a geiger counter x-ray spectrometer," J. Appl. Phys., Vol. 20, p. 1030, 1949.

stereographic diagram is essentially the same as Figure 42. For sheet material the reflecting planes are assumed to be in a plane representing the sheet which is parallel to the Debye ring plane and the projection plane. This plane is midway between the two planes shown and contains the center of the reflection sphere which is actually the point of diffraction. Therefore, the angle the plane makes with the rolling plane, the plane of the sheet, now becomes the angle away from the center of the pole figure. The angle the plane makes around the axis of the x-ray beam, now becomes the angle around a axis normal to the sheet itself. In that the plane has a rolling direction which can serve as an axis of reference in the rolling plane itself, it is possible to define the α angle with respect to the pole figure and the rolling direction. The rolling direction is perpendicular to the x-ray beam and is normally considered to be on the top of the projection plane. As an illustration of how this relationship would work, a reflection plane normal to the rolling plane would be represented by a point somewhere along the outer edge of the projection circle - the periphery of the pole figure. If the plane was not only normal to the rolling plane but also normal to the rolling direction, the pole through the reflection plane would make points at the extreme top and bottom of the outer edge of the pole figure.

The density of the projected pole points in a particular area of the pole figure is a measure of the preferred orientation of the sample in question. In practice this point density is found by measuring the intensity of the diffracted radiation of the planes that

correspond to a particular orientation. The technique involved is to set the x-ray diffraction spectrometer for a 2θ angle corresponding to the Bragg reflection plane of interest, and then move the sample through various orientations with respect to the projection plane. High intensities correspond to a great number of poles being oriented in a similar manner, thereby indicating some degree of preferred orientation.

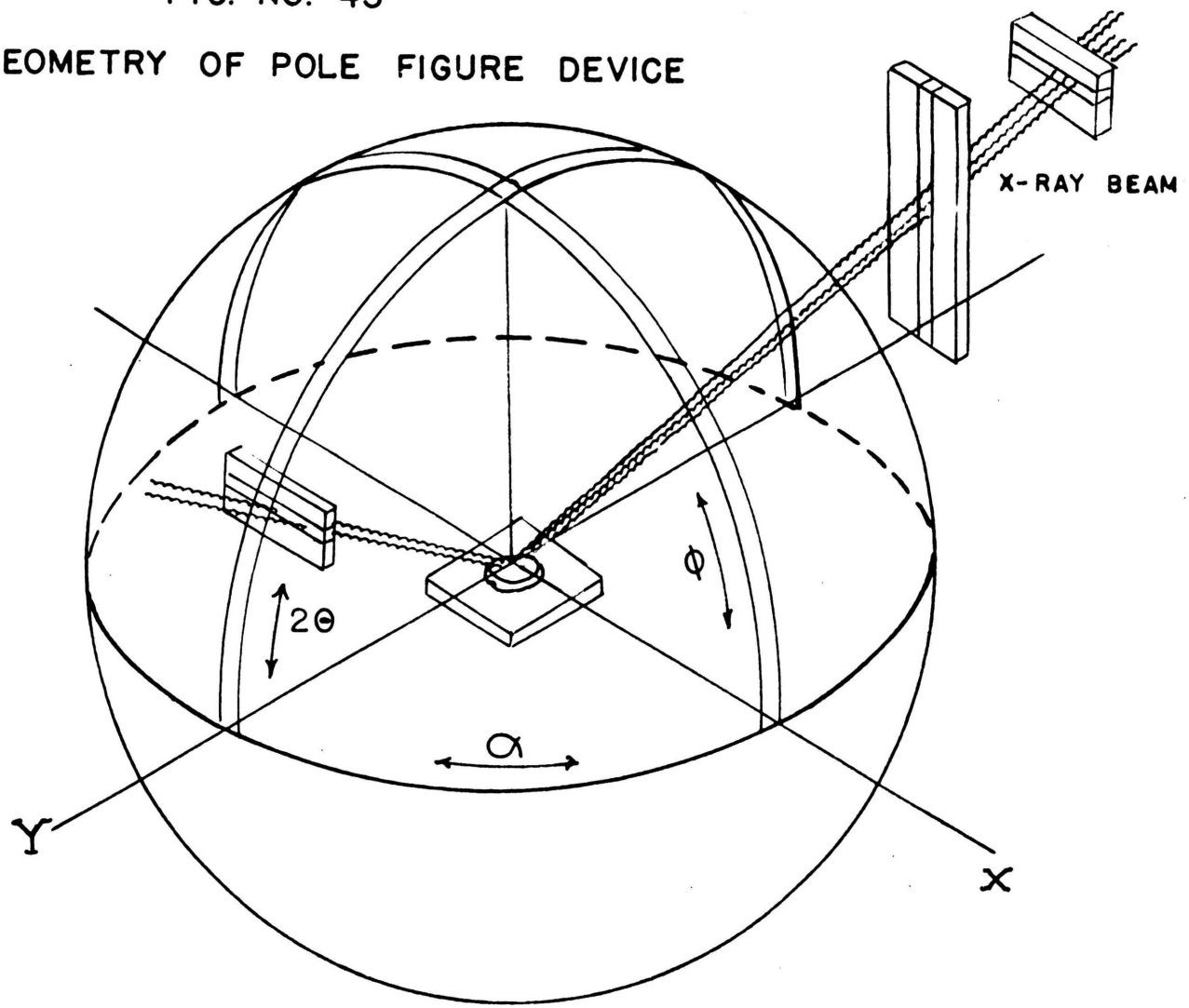
B. REFLECTION TECHNIQUE FOR TEXTURE DETERMINATION OF SHEET SAMPLES

Geometry of pole figure device. The original Schultz technique has been refined in recent years through the development of automatic specimen holders. The use of the x-ray diffraction spectrometer with Geiger counter has remained essentially unchanged. The geometry of the basic pole figure device is shown in Figure No. 43. In this discussion, as in Figure 43, Norelco geometry is assumed; the sample is horizontal with the diffraction and the 2θ rotation being in a vertical plane about the χ axis. It could be considered for horizontal type diffractometers by rotation of the diagram.

The sheet sample is placed on the circular stage in the center of the reflection sphere shown in Figure No. 43. In this position horizontal rotational provides α movement while rotation around the Y axis provides ϕ movement. It is immediately obvious that the sample itself will absorb the x-ray beam when ϕ rotation approaches the vertical position. The X axis is also the rotational axis of the x-ray diffractometer. The x-ray beam originates at the right hand

FIG. NO. 43

GEOMETRY OF POLE FIGURE DEVICE

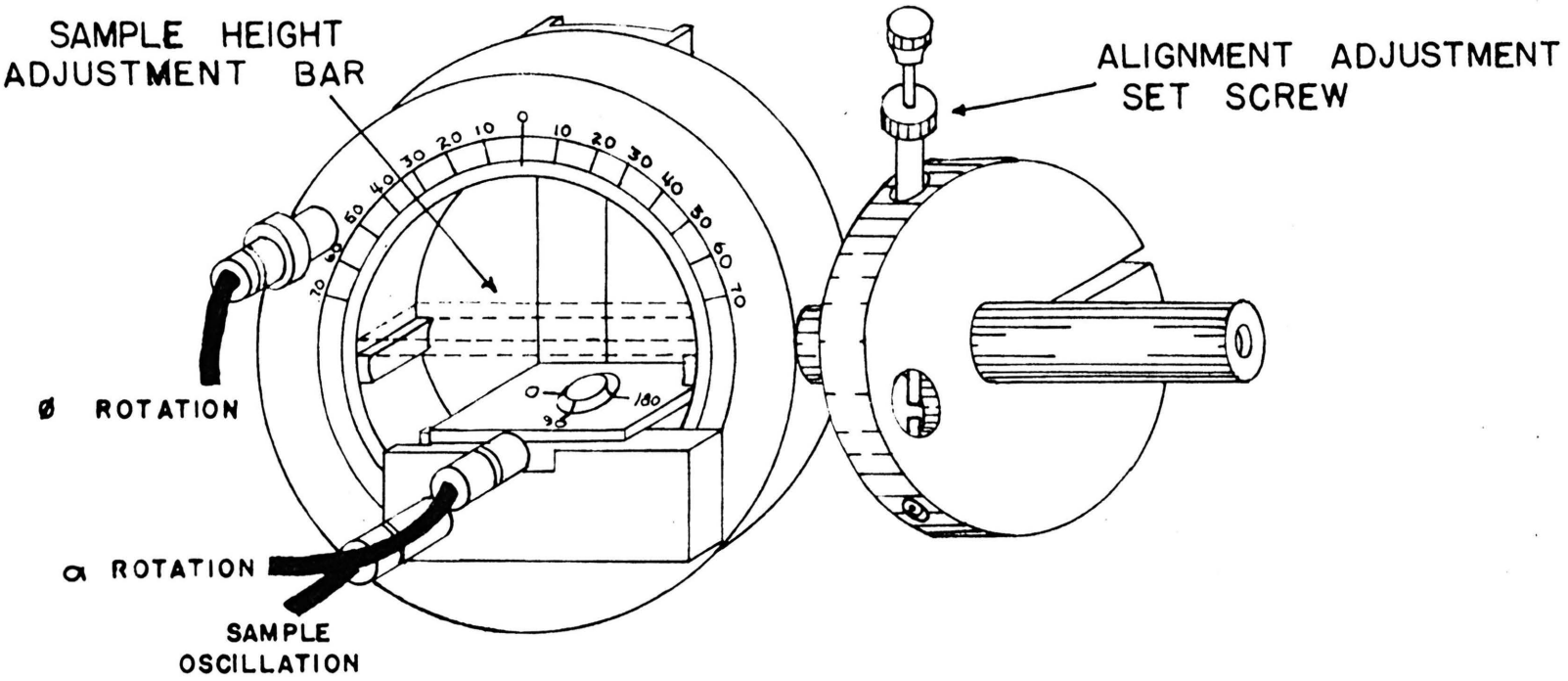


side of the diagram, is collimated, diffracted by the sample and recorded by a Geiger tube at the left hand side of the reflection sphere. In practice, the diffraction geometry is established and the tube position not changed during a particular determination. The sample is rotated through α and or ϕ arcs either in steps or continuously, the diffracted intensity for the particular plane of interest being recorded. By this method the inner 60° portion of the pole figure can be covered.

The Norelco fixture. The pole figure fixture developed by the Philips Electronic Division of the North American Philips Company incorporates the geometry illustrated in Figure No. 43. The fixture itself is illustrated in Figures 44, 45 and 46. Flexible drive shafts coupled to motors in an external cabinet provide appropriate sample movement. The outer ring, ϕ rotation, is designed to be driven in either direction of rotation at a rate of 1° per minute. The specimen table can be rotated at a rate of 10° or 20° per minute, thus providing continuous movement. The specimen table is made to oscillate in its own plane at a rate of one oscillation per second. By exchange of eccentric drive cams the distance of the oscillation stroke can be set for 1/16", 1/4" or 3/8". All three drives are independent of each other and can be operated separately or in any combination.

Alignment of the fixture. Accurate alignment of the pole figure fixture can be obtained by basically the same techniques used for a regular powdered sample holder. With the vertical slits on the tube side of the fixture removed, the stage is elevated so that the

FIG. NO. 44



NORELCO POLE FIGURE FIXTURE

FIG. NO. 45

DISSASSEMBLED VIEW OF NORELCO POLE FIGURE FIXTURE

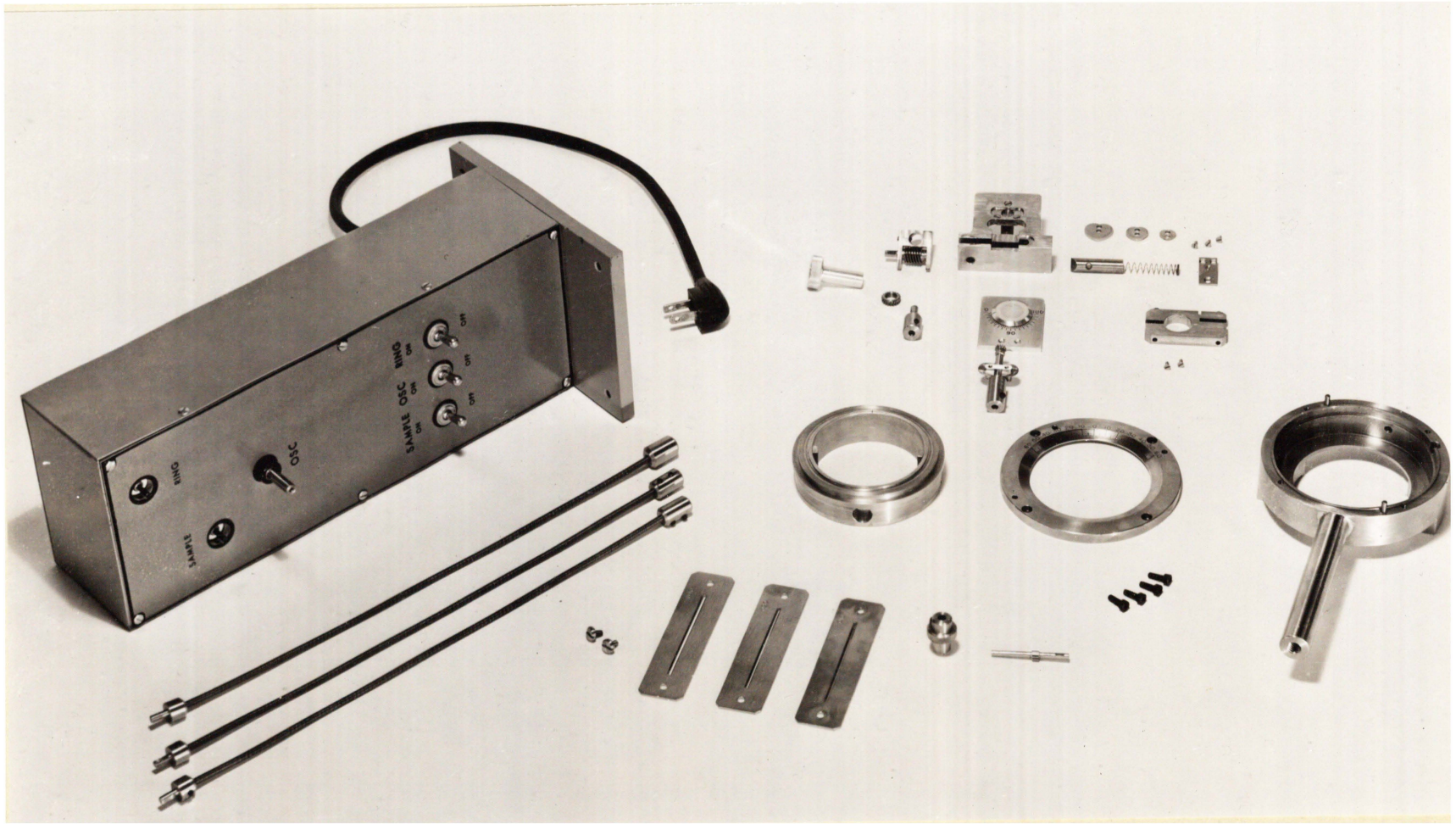
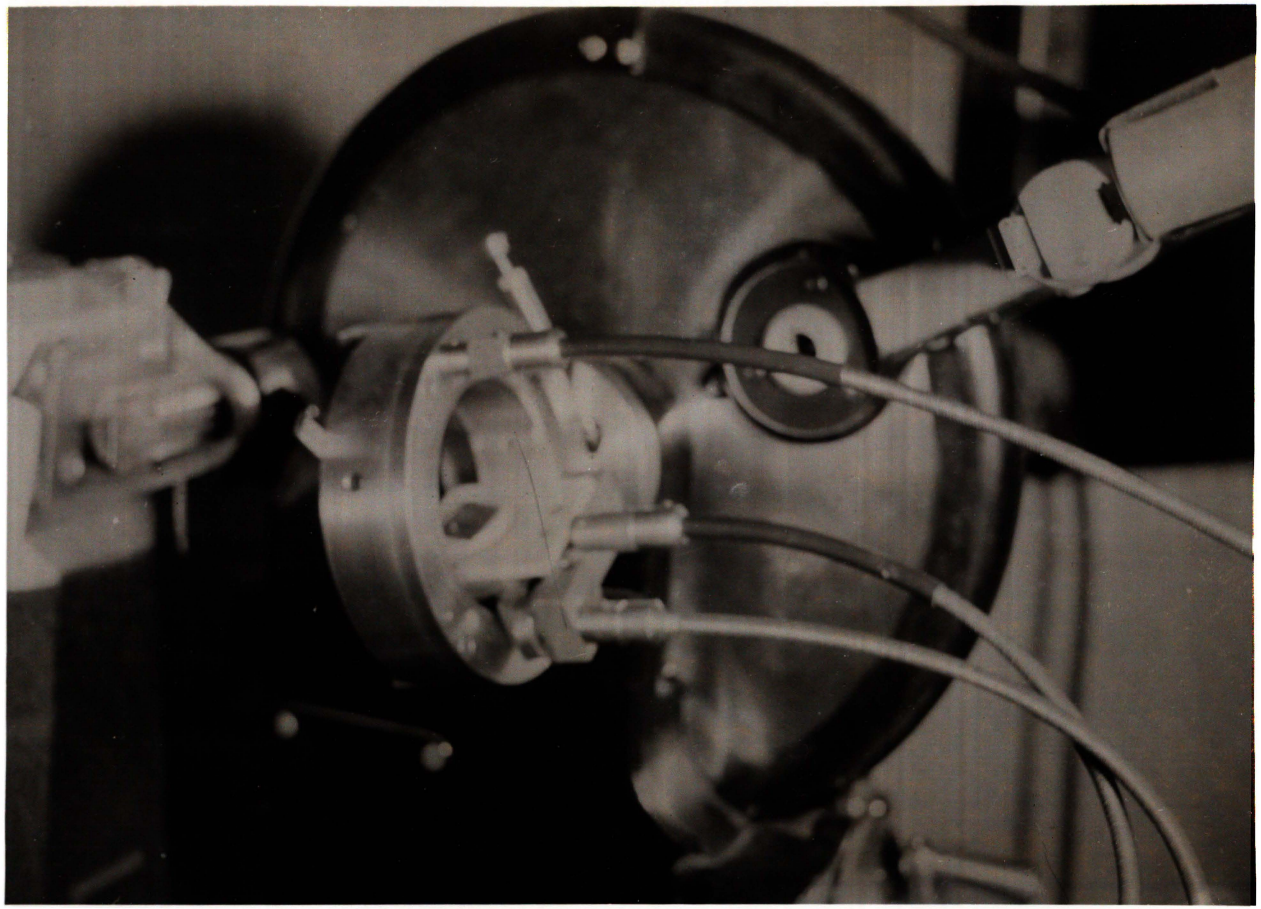


FIG. NO. 46

NORELCO POLE FIGURE FIXTURE IN OPERATION



axis of the diffractometer runs through the stage surface and the ϕ setting at 0°, the procedure outlined in the Norelco Instruction manual can be followed (97).

The sequence of this procedure, as modified for the pole figure fixture, is to first align the diffractometer so that the axis of the goniometer is at the desired take-off angle. This is done by adjusting the height of the axis of rotation of the goniometer with respect to the plane of the tube anode. The normal take-off angle of 3° was used for all the work described in this investigation. This angle appears to give satisfactory intensity. If necessary the intensity may be increased, at the expense of resolution, by a slightly larger take-off angle. Take-off angles up to 6° can be used.

The axis of rotation of the goniometer must be placed parallel to the long axis of the local spot. This involves positioning the goniometer as nearly parallel as is possible to the face of the tube.

After this positioning of the diffractometer has been made as close as possible for the desired take-off angle, the alignment bar can be used to adjust the mechanical zero of the goniometer. The measuring dial of the goniometer is set for 0°. The surface of the stage of the sample holder is elevated so that it contains the axis of rotation of the goniometer. This can be done by running the stage up until it just meets the horizontal height adjustment bar shown in in Figure 44. The alignment bar is then held firmly against the stage

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(97) "Instruction and Operating Manual - Wide Range Goniometer," North American Philips Company, New York.

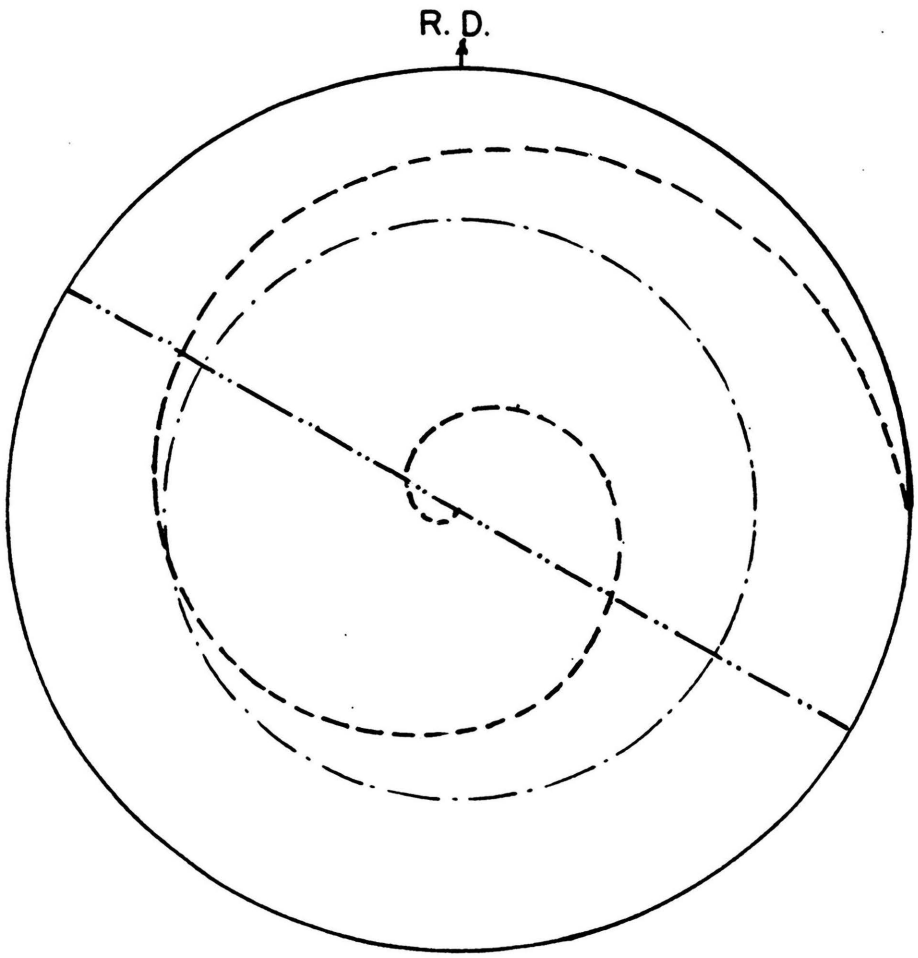
FIG. NO. 47

POLE FIGURE SCANNING PROCEDURES

—··—··— = Ø ROTATION

—·—·—·— = α ROTATION

— — — — = SPIRAL SCAN, 10° α PER 1° Ø



and by means of the alignment set screw the fixture is adjusted so that the points of the alignment bar bisect the divergence and receiving slits. The fixture head is then tightened.

This approximate alignment can be refined by placing the fluorescent alignment screen on the fixture stage and with the use of a relatively soft x-ray beam checking the position of the beam image. The height of the stage and sample must be set with the height adjustment bar so that the image contains the rotation axis of the goniometer. The fixture should be rotated to check that position of the image on the screen is coincident with the axis of rotation of the goniometer. It may be necessary at this point position the goniometer slightly to bring the axis of rotation of the goniometer parallel to the axis of the image. Leveling of the goniometer may also be required which can be accomplished by adjustment of the three leveling screws on the base of the diffractometer.

The alignment of the fixture can be easily checked by running a standard silicon pattern. More refined alignment procedures are suggested by the manufacturer (98) but generally speaking the alignment precision required for pole figure work need not be as precise as most other work. The procedure outlined above which parallels the procedure outlined for powder specimen holder in the goniometer instruction manual is usually entirely satisfactory.

Sample preparation. It is desirable to prepare a sample in such a manner that the texture recorded is representative of the material.

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(98) Chernock and Wahl, Loc. cit.

In transmission work this envolves no problem, but in reflection determinations the surface of the sample must be carefully prepared. Braybrook and Calnan note that at least 0.001" should be removed from the surface and this should be removed in such a manner that the sample is free of distrubed metal resulting from mechanical polishing. The best procedure is to use mechanical polishing followed by chemical etching. An etchant should be used which will leave a bright surface free of etch products as was obtained with 10% HF solution with hafnium. Care should be taken to keep the sample flat without bending or stressing it. This sometimes involves polishing and etching both sides to keep a thin sample from warping.

Alignment of sheet sample on fixture stage. The sheet sample for texture analysis must be as flat as possible. Samples up to 5/16" thick and up to 1" in diagonal or diameter can be accommodated on the fixture. The samples can be affixed to the stage with rubber cement or gasket cement. The rolling direction of a cold rolled sample should be made to coincide with the reference marks on the beveled edge of the stage. The stage can then be elevated until the sample comes in contact with the horizontal height adjustment bar. The bar should be held firmly against the side lugs and care should be taken so that the sample is not raised so much that the bar is lifted.

The sample oscillation should be adjusted for the size of the sample. The entire beam should fall on the sample in any position and small samples therefore require the shortest oscillation distance.

After the sample is positioned it is desirable to make a 2θ

scan so that the 2θ angle for the particular Bragg reflection to be investigated can be set with maximum accuracy. In highly oriented samples it is sometimes necessary to adjust the ϕ and α positions to get maximum peak heights in the 2θ scan. After the desired 2θ angle has been determined, it can be set on the goniometer and need not be changed or altered during an entire pole-figure run. Very small changes in 2θ can cause intensity variations.

α and ϕ scanning. Once the diffraction geometry for a given reflection is set, the α and ϕ movements can be actuated to move the sample in such a manner as to cover the desired area or areas of the pole figure. The two movements can be used separately or together and these three possibilities are shown graphically in Figure 47. As can be noted if one uses ϕ scan, a series of intensity values constitute a straight across the pole figure. The whole pole figure can be covered by manually readjusting the α angle after each scan and making another ϕ scan. This was the principle employed by many of the earlier units. The intensity of coverage of the pole figure is determined by how closely in α angle the ϕ scans are run.

A second possibility is to motorize α drive only, after setting the ϕ angle at the desired position. Selective pole figure circles can be made by resetting the ϕ angle after each complete rotation. This has an advantage in that particular areas can be covered with any desired density of determinations. This is particularly useful where all four pole figure quadrants are being run and when the exact position of the peak of maximum intensity is desired.

The density of readings can be reduced by making larger ϕ increments where no peaks are known to exist. This is the general procedure used in this investigation. Complete α circles were made at 5° ϕ increments in areas where peaks occurred.

A combination of α and ϕ scans, spiral scanning as it is called, gives good general continuous coverage of the entire pole figure without any readjustment. The coverage or density of readings can be varied by choosing the appropriate combination of motor drives. It is also possible to get dense coverage by running a spiral scan as shown in Figure 47; then turning the sample a given α angle and running another spiral scan. The single scan shown in Figure 47 represents 10° α rotation per 1° ϕ rotation. A greater density of readings is possible by using the alternate 20° α rotation per 1° ϕ rotation.

C. CORRECTION OF EXPERIMENTALLY DETERMINED INTENSITY VALUES

Tilting angle correction. As has been pointed out by Chernock and Beck[99], tilting angle correction adjustments are necessary to get accurate intensity values. It has been found there is a general attenuation of the diffracted beam with increasing ϕ angle. This is due to the collimated incident beam falling on an increasingly larger area thereby cutting down the intensity of the collimated diffracted beam. Very small alignment errors can also cause slight additions to the tilting angle correction necessary. The tilting angle correction

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(99) W. P. Chernock and P. A. Beck, Loc. cit.

necessary at the center area of the pole figure is insignificant but amounts to as much as a factor of four at ϕ equals 60°.

Tilting angle corrections are made by preparing a powder sample of the metal to be examined. A powder should be employed that will give a randomly oriented specimen. The powder used in this investigation was -400 mesh annealed hafnium filings. The sample is mounted on the specimen stage and the variation of intensity with changing ϕ angle is recorded. It is well to use the sample scanning procedure proposed for the subsequent investigation. The correction factor calculated for a specific ϕ angle is the multiplying factor necessary to make the intensity at that angle in the random sample equal to the intensity of ϕ equals 0°. A tilting angle correction chart for hafnium powder which was used in this investigation is shown in Figure No. 48.

Counting efficiency. In normal pole figure work the intensity corresponding to orientation peaks is sometimes as much as 30 to 40 times as much as the areas where no distinct peaks occur. It is desirable that the absolute intensity values be comparable over this whole counting region. For this reason it is necessary to know the counting rate at which the efficiency of the Geiger tube begins to fall off. This variation from linearity can be determined by placing a series of calibrating filters in the diffracted beam and removing them one at a time. The increase in counting rate for such a determination is shown in Figure 49. At approximately 600 counts per second there is a deviation from linearity. In order to get absolute intensity values in this investigation a slit system was used which

kept the intensity below 600 counts per second and so no correction factor was necessary. Had it been necessary to go above that intensity a correction factor could have been calculated for any particular intensity. In this investigation a 1° divergence slit, a 0.20 vertical fixture slit and a 4° tube slit were employed.

FIG. NO. 48

TILTING ANGLE CORRECTION FACTOR

MULTIPLYING FACTOR BASED ON ATTENUATION OF COPPER K_α RADIATION DIFFRACTED FROM POWDERED HAFNIUM SAMPLE

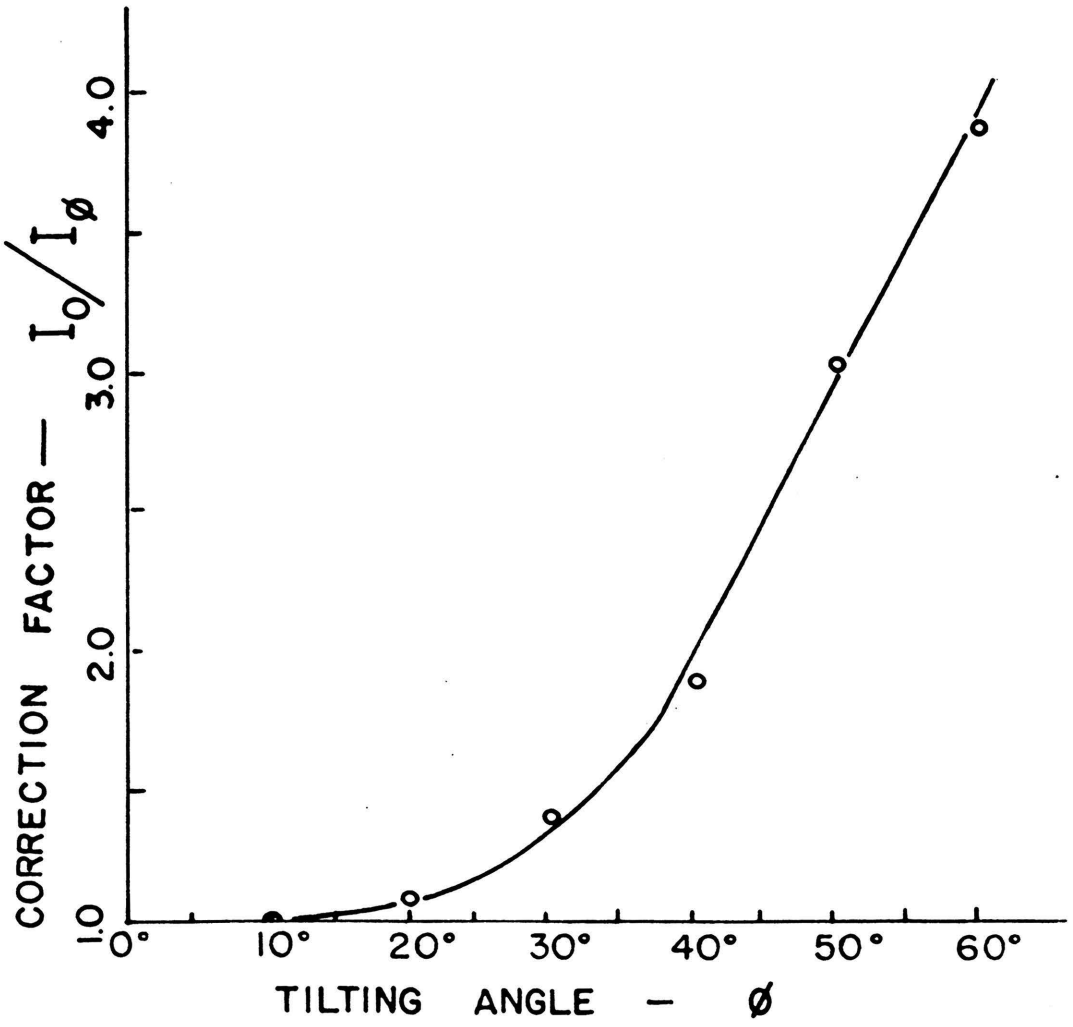
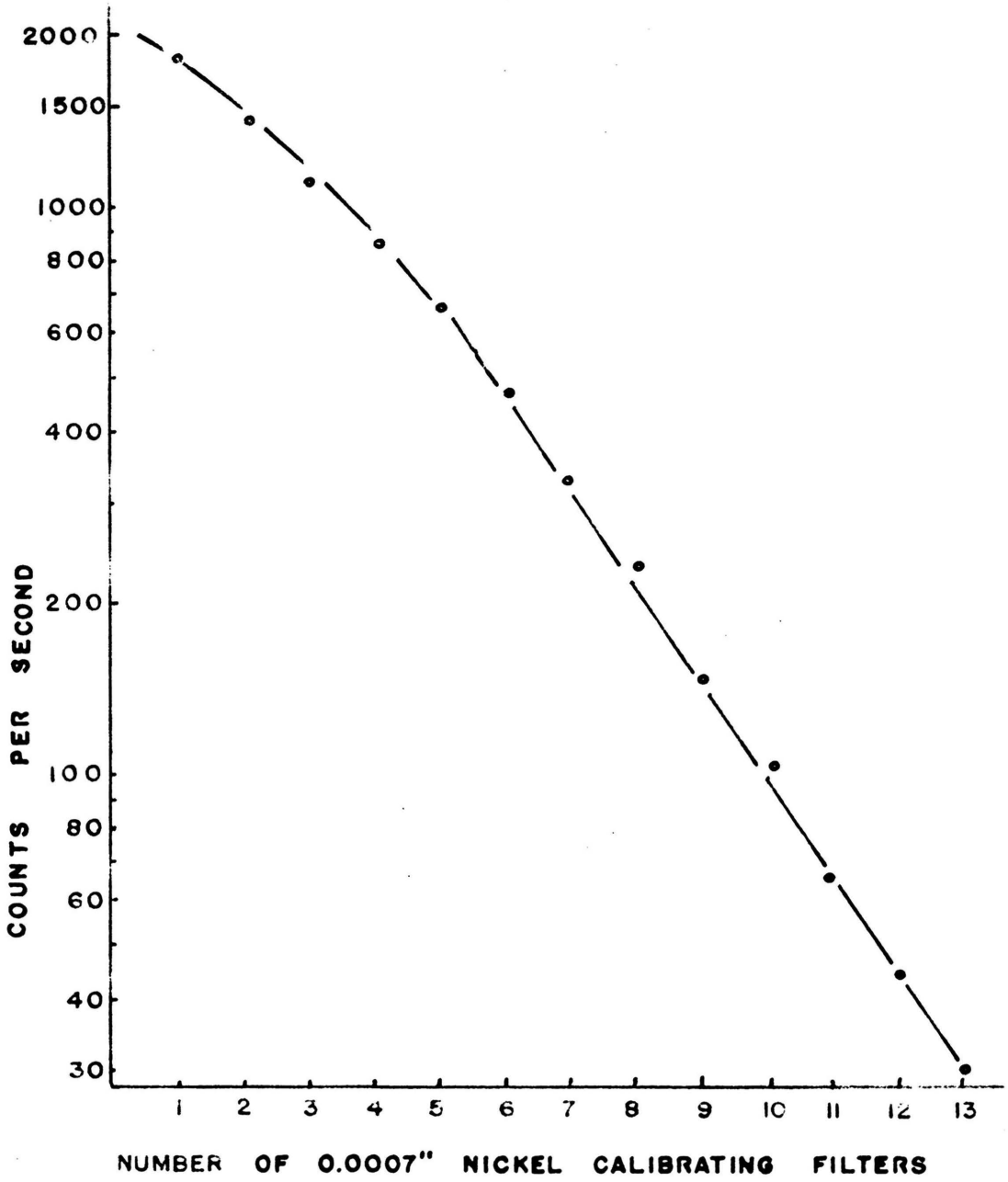


FIG. NO. 49

GEIGER TUBE LINEARITY DETERMINATION

TUBE NO. 10564 OPERATING AT 1575 VOLTS COUNTING COPPER K_α RADIATION.



VITA

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